

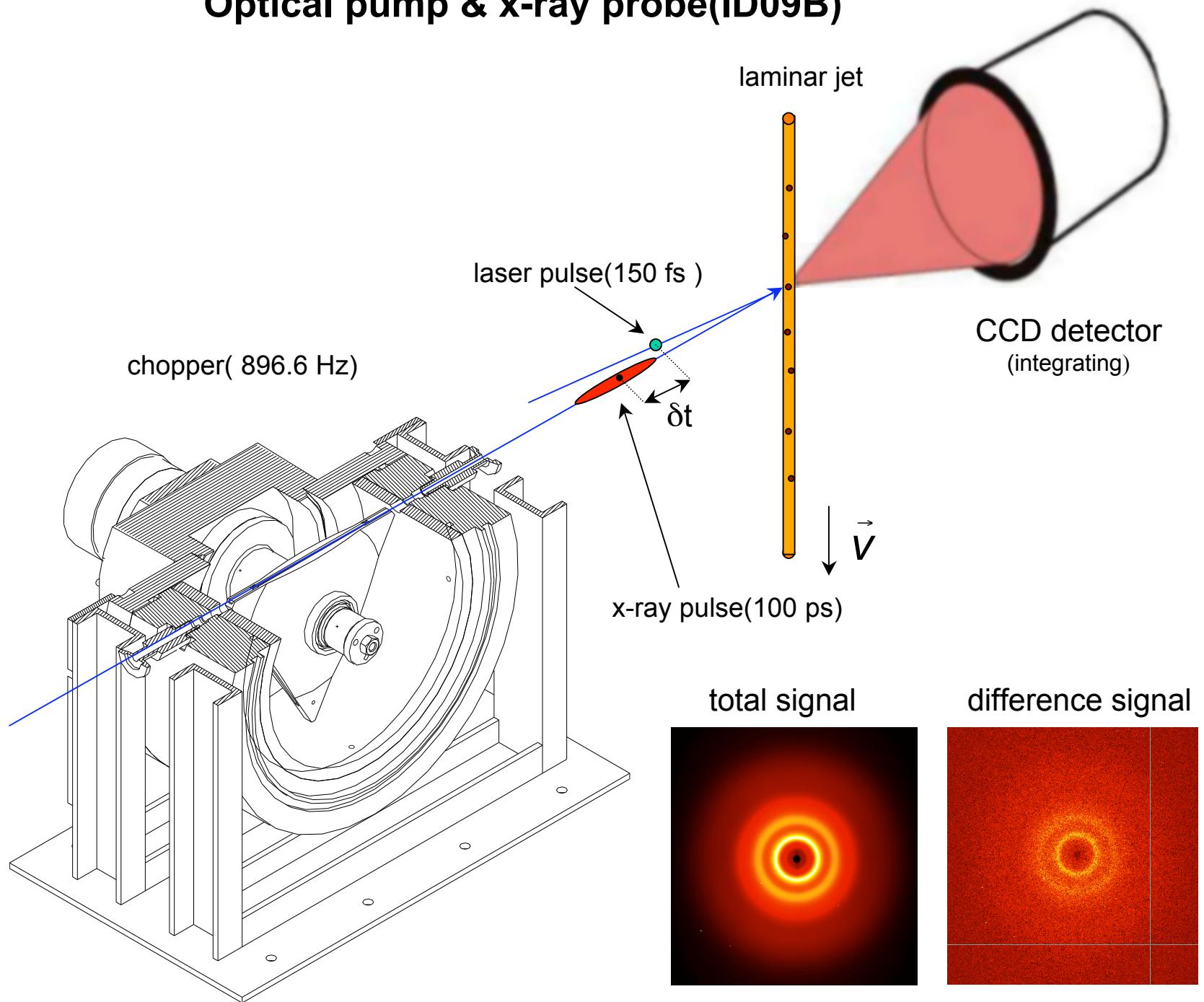
Time-resolved studies of molecular structure with pulsed x-rays:
where are we and where are we going?:

Michael Wulff

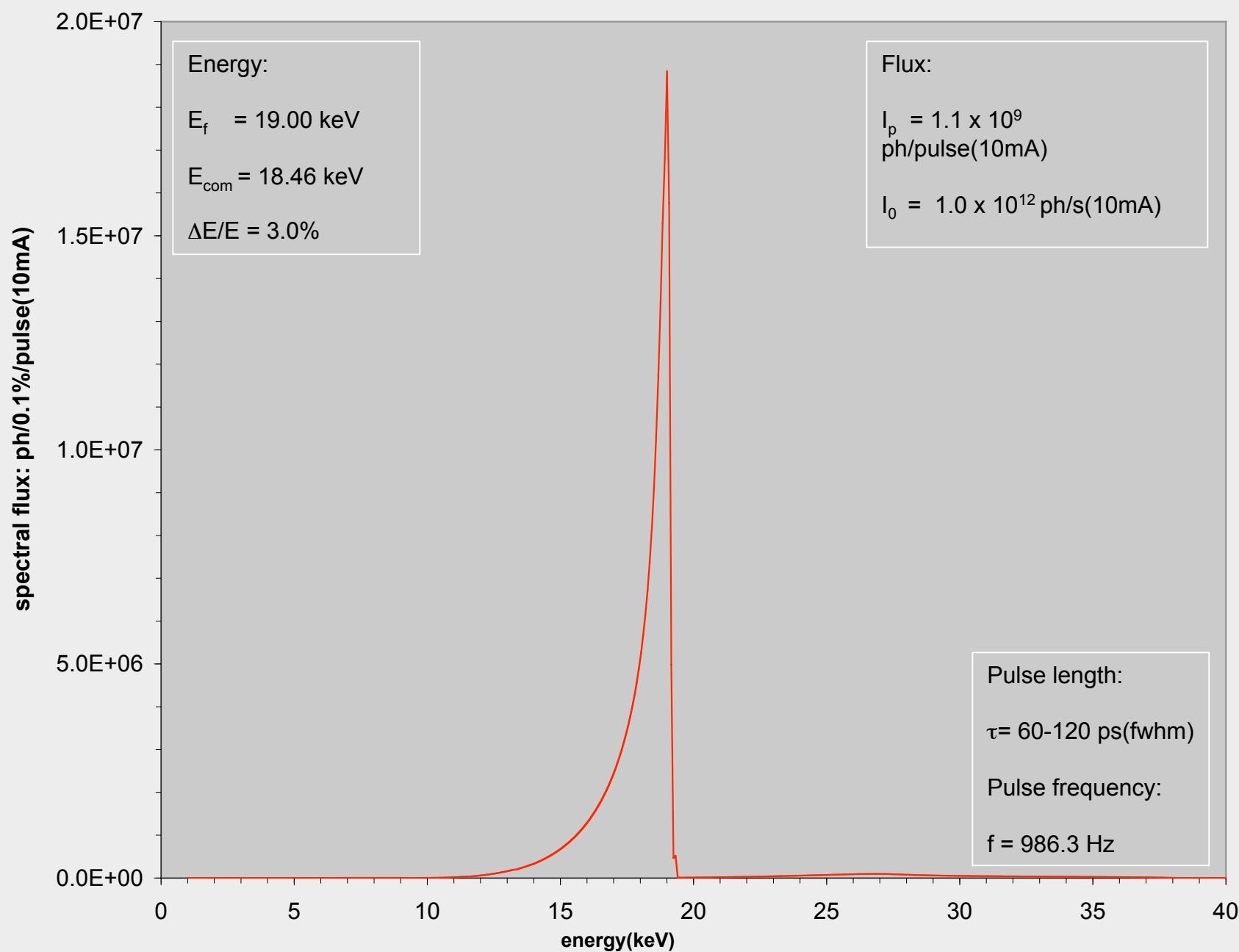
European Synchrotron Radiation Facility, Grenoble, Cedex 38043, France



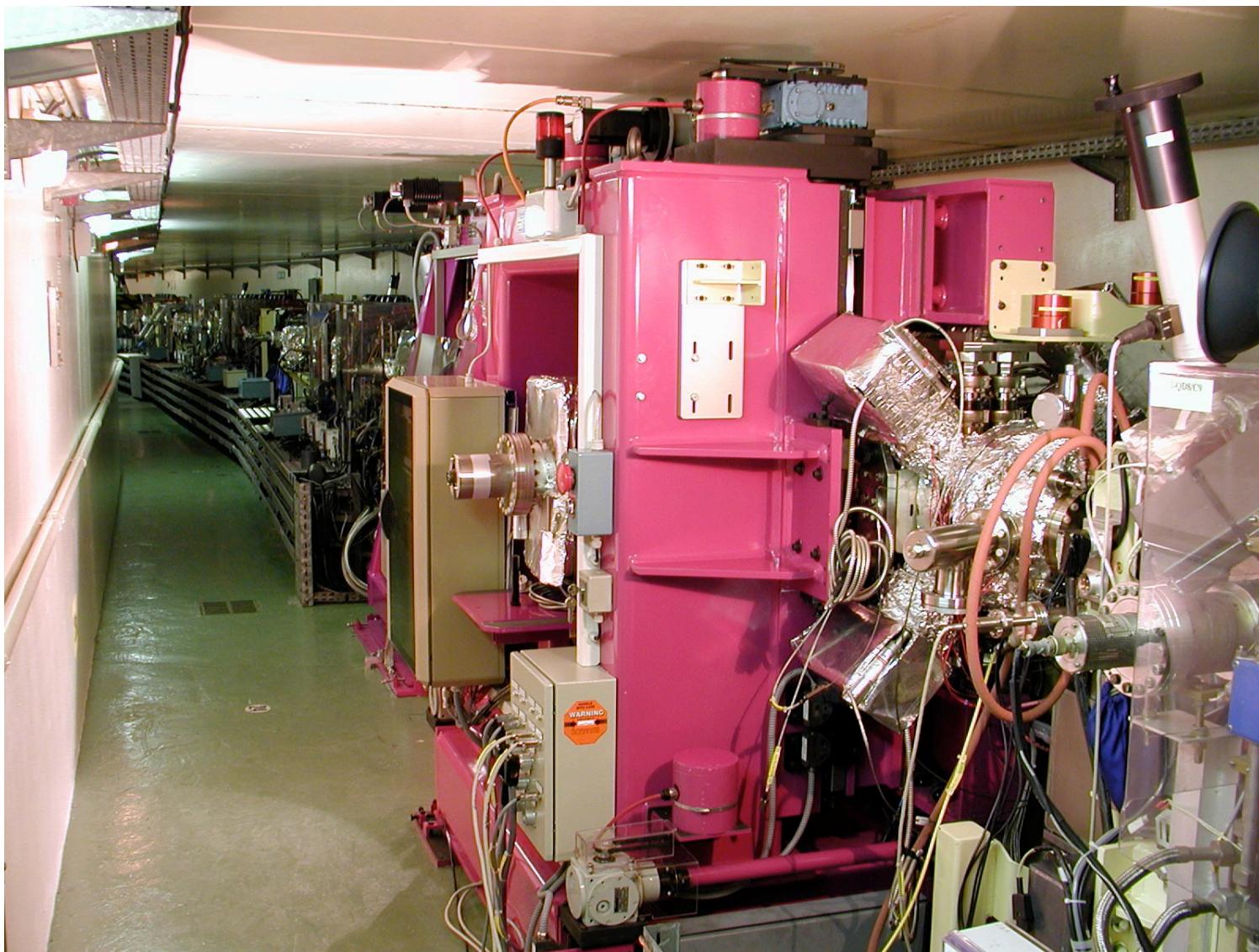
Optical pump & x-ray probe(ID09B)



Spectrum of the mono-harmonic undulator U17



The U17 undulator on ID09



IDpolesgap (mm)	E ^f (keV)	E ^c (keV)	KP(W/200 mA)	U172356.014.8413.20.862740		
					400	

The structural dynamics of small molecules in liquids I_2 , Br_2 and $C_2H_4I_2$

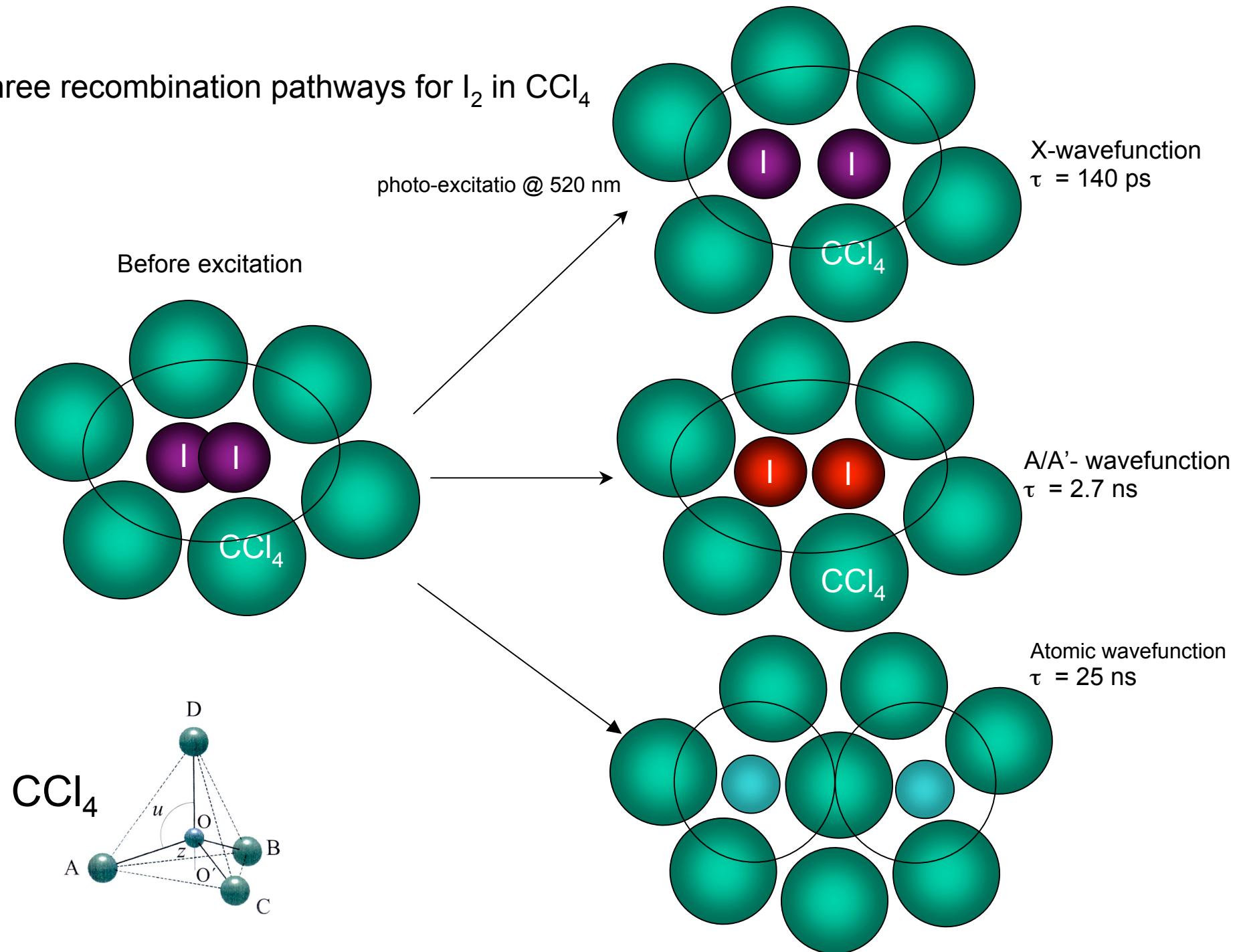
Experiment:

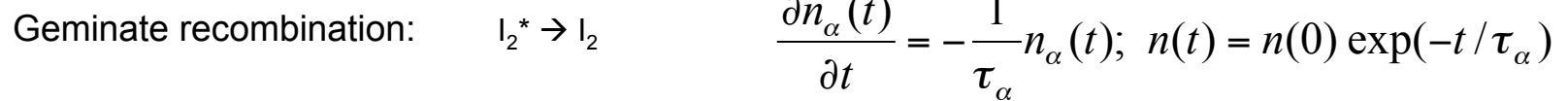
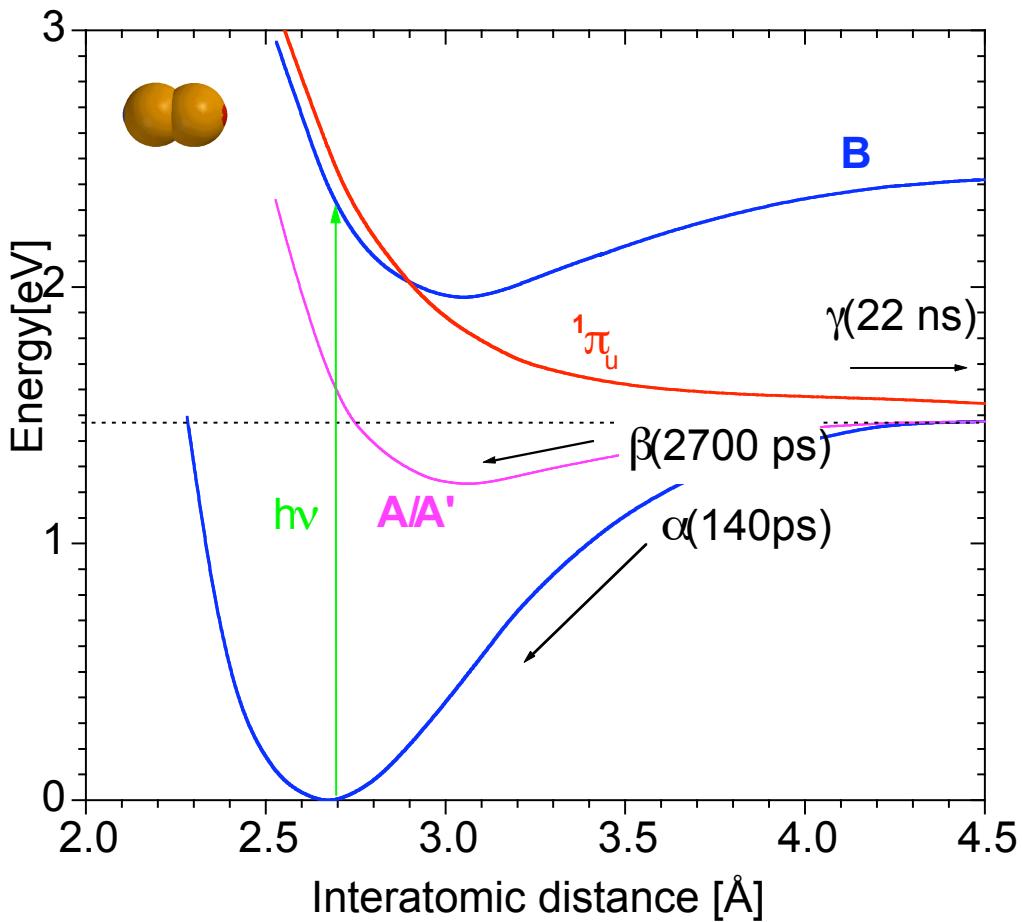
Maciej Lorenc, Qingyu Kong, Hyotcherl Ihee, Anton Plech, Philip Anfinrud, Friedrich Schotte and Michael Wulff,

Theory:

Fabien Mirloup, Rodolphe Vuilleumier, Savo Bratos

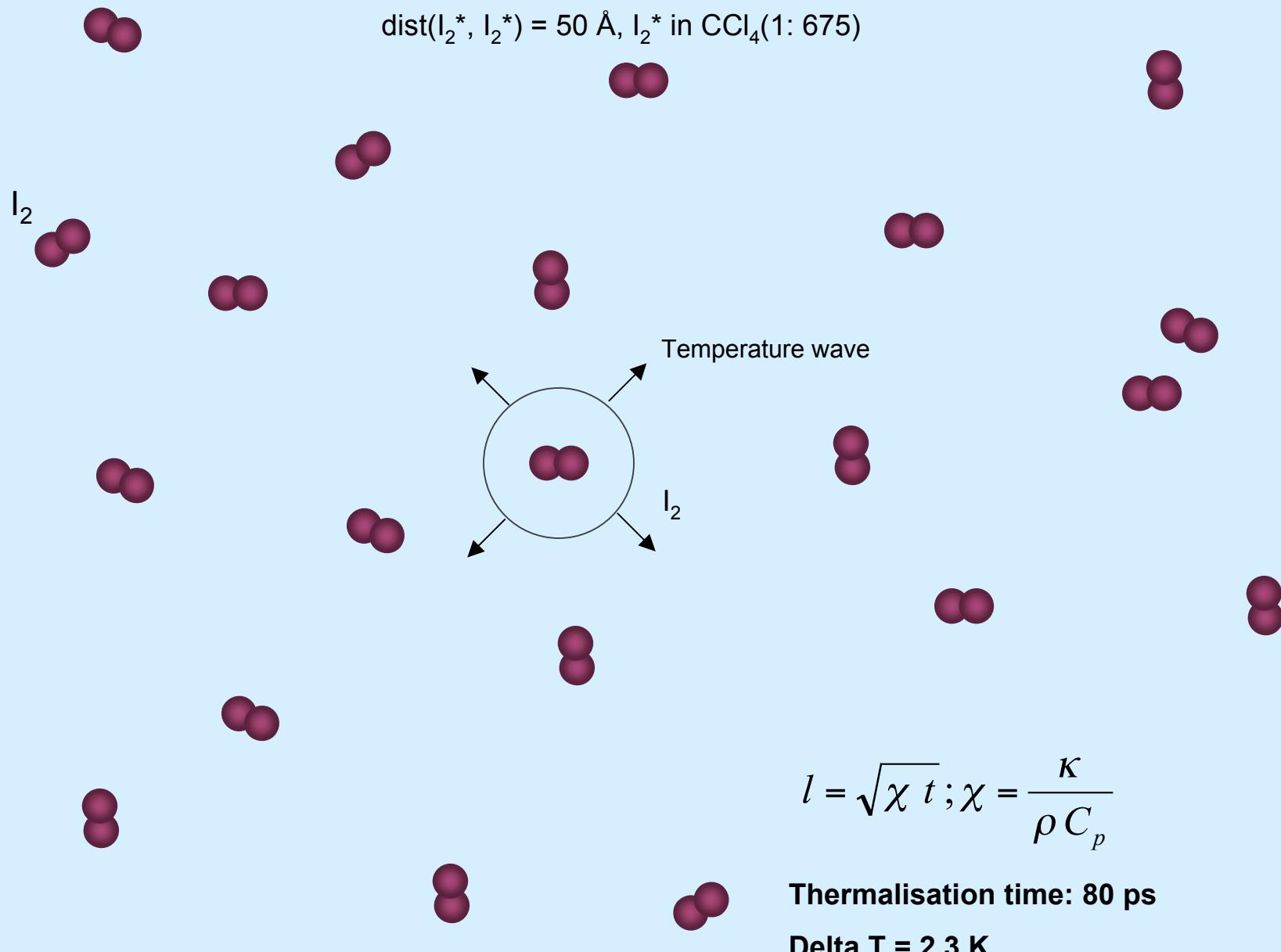
Three recombination pathways for I₂ in CCl₄



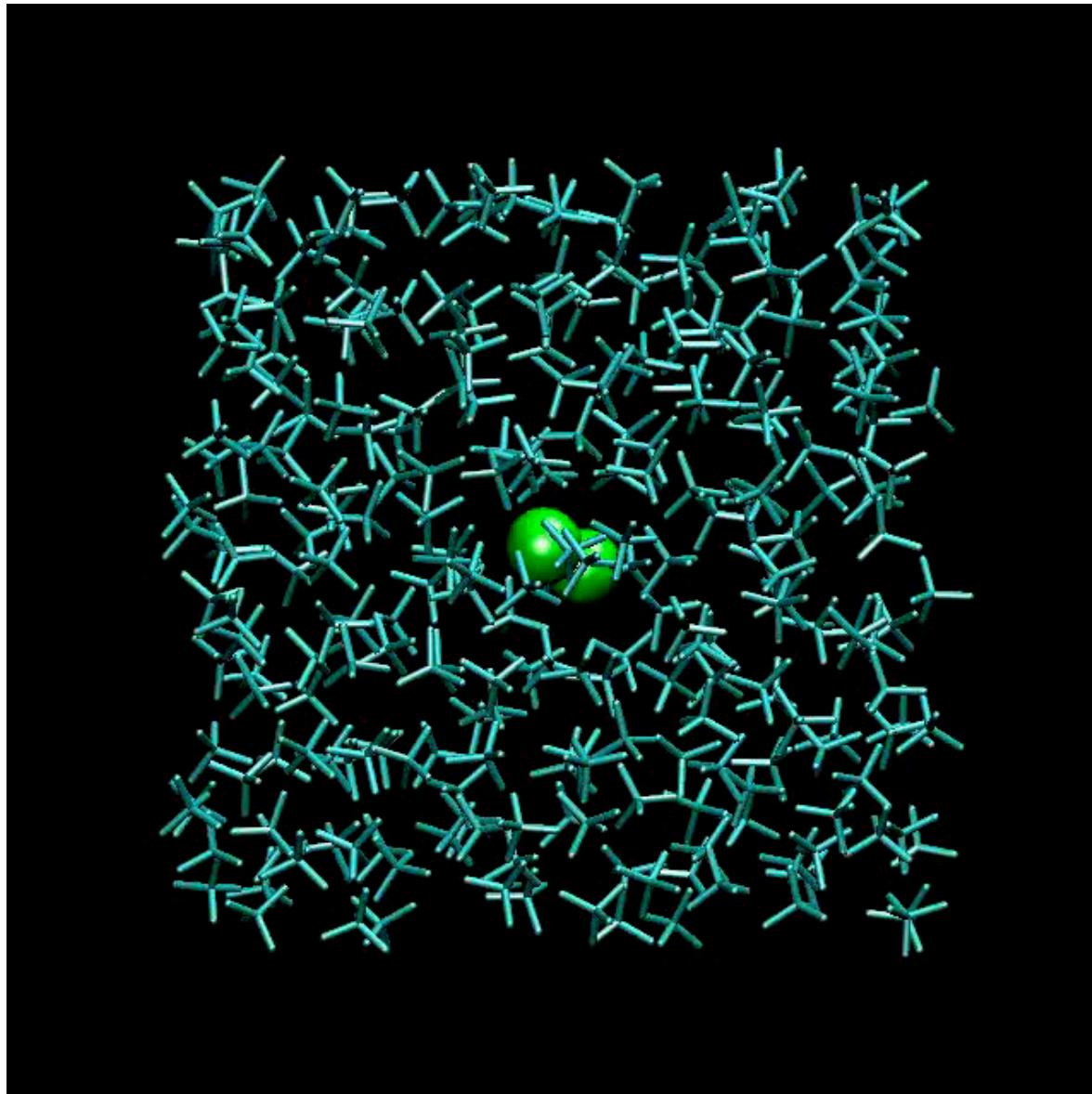


$$k_\gamma = 0.72 \cdot 10^{-2} \text{ M}^{-1} \text{ ps}^{-1}$$

The time to reach local thermal equilibrium in CCl_4

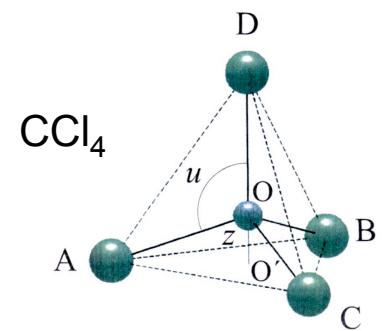


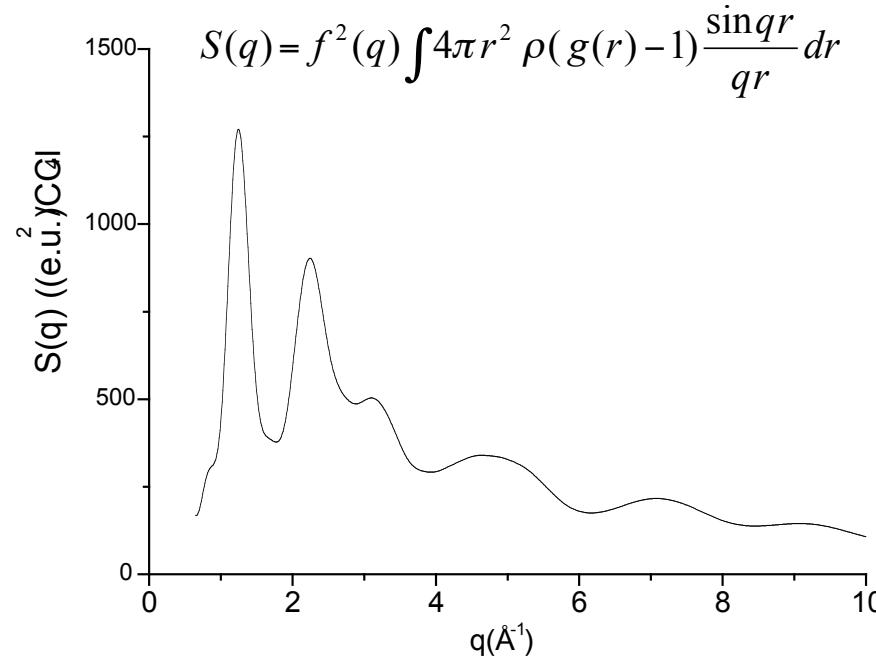
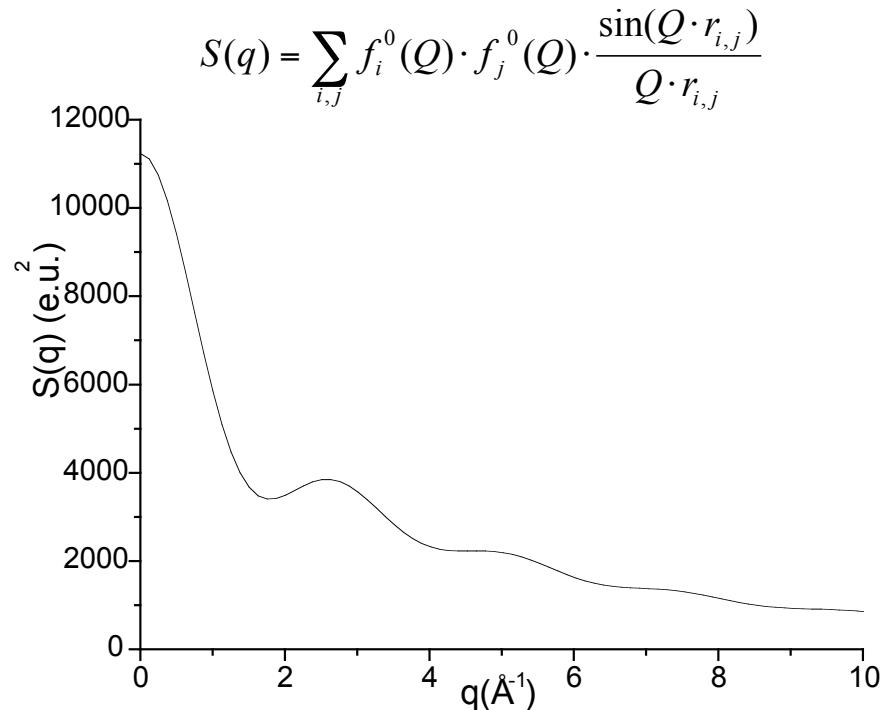
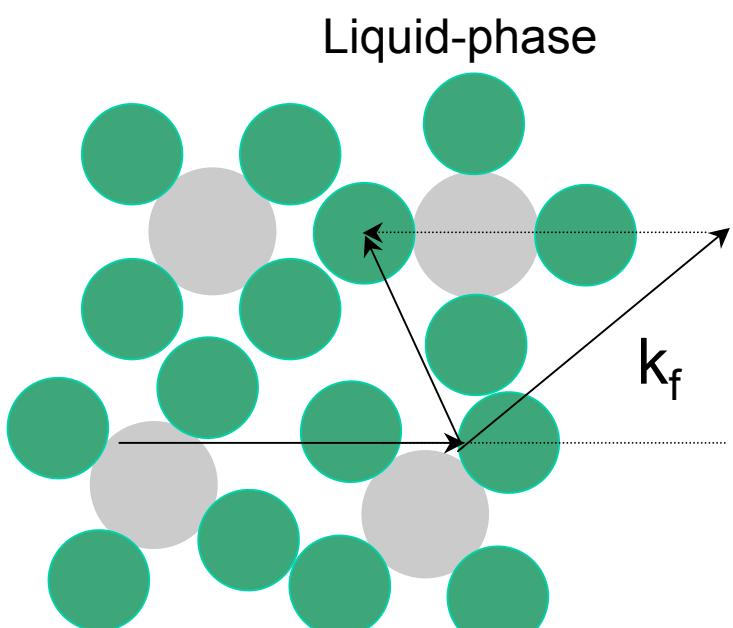
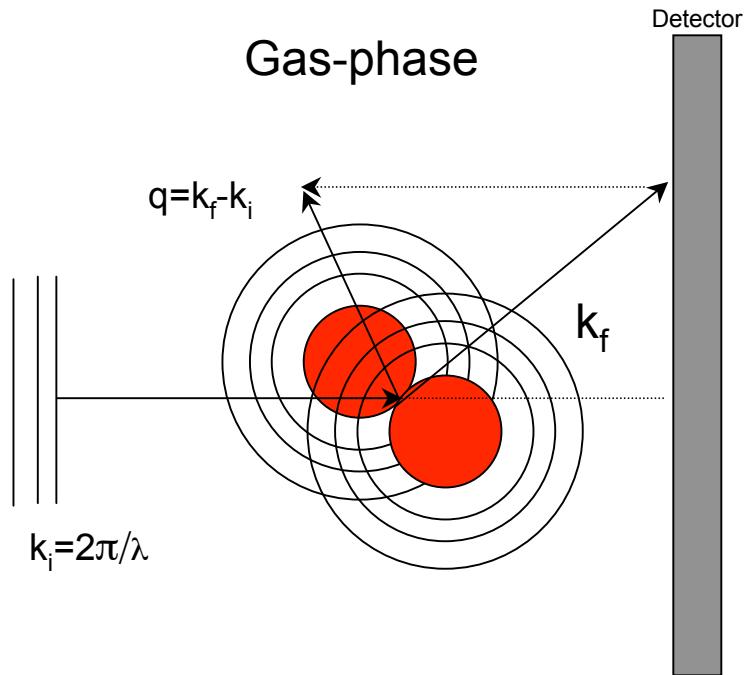
The dissociation and recombination of I₂ in CCl₄



MD simulation by Rodolphe Vuilleumier, Paris(CP2K)

256 CCl₄ + I₂. Time-step 0.5 fs(20 fs steps shown here). Film stops at 10 ps



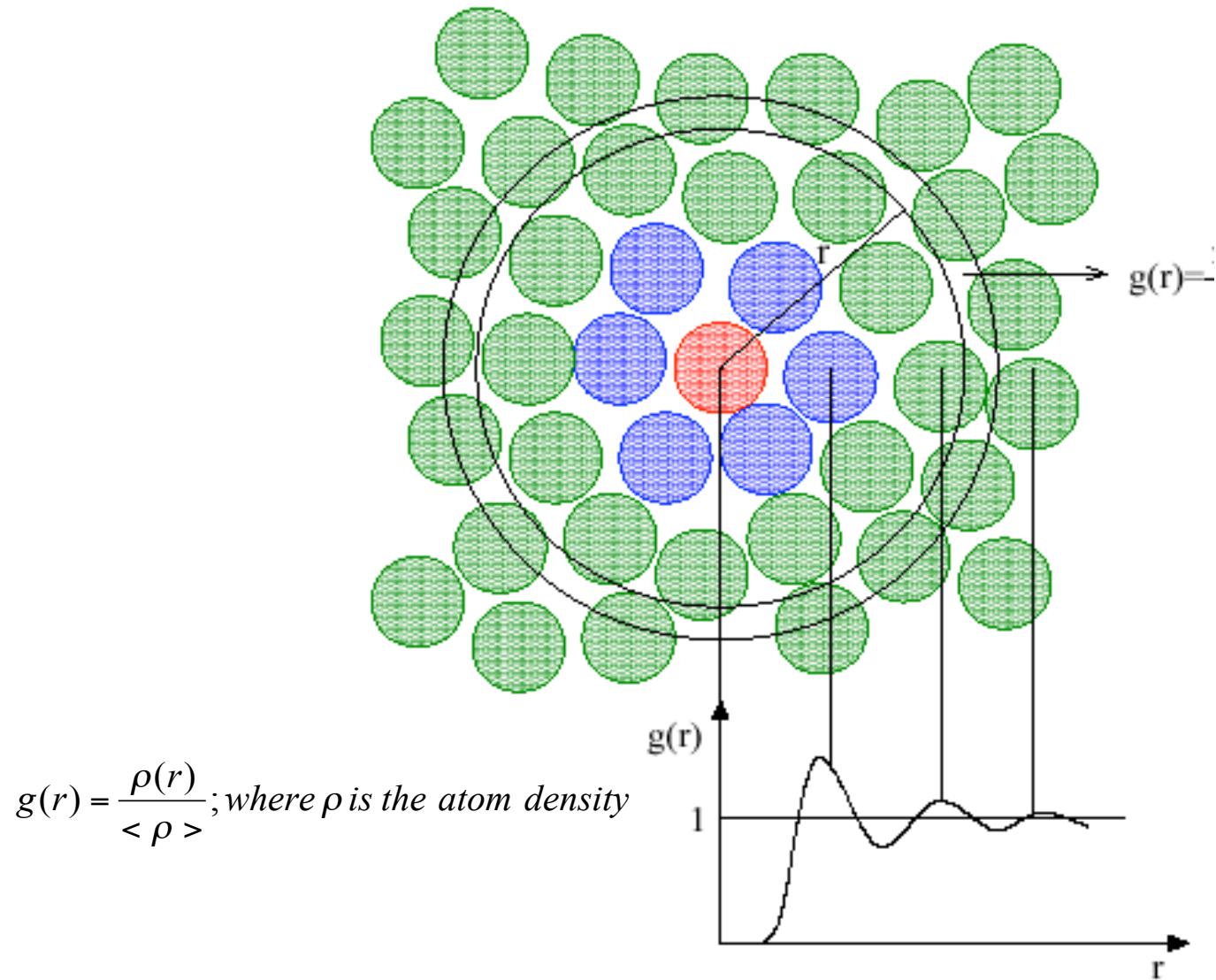


Real-space inversion of $\Delta S(q, t)$ to $\Delta S[r, t]$

$$\Delta S[r, \tau] \equiv \frac{1}{2\pi^2 r} \int_0^\infty dq q \Delta S(q, \tau) \sin(qr)$$

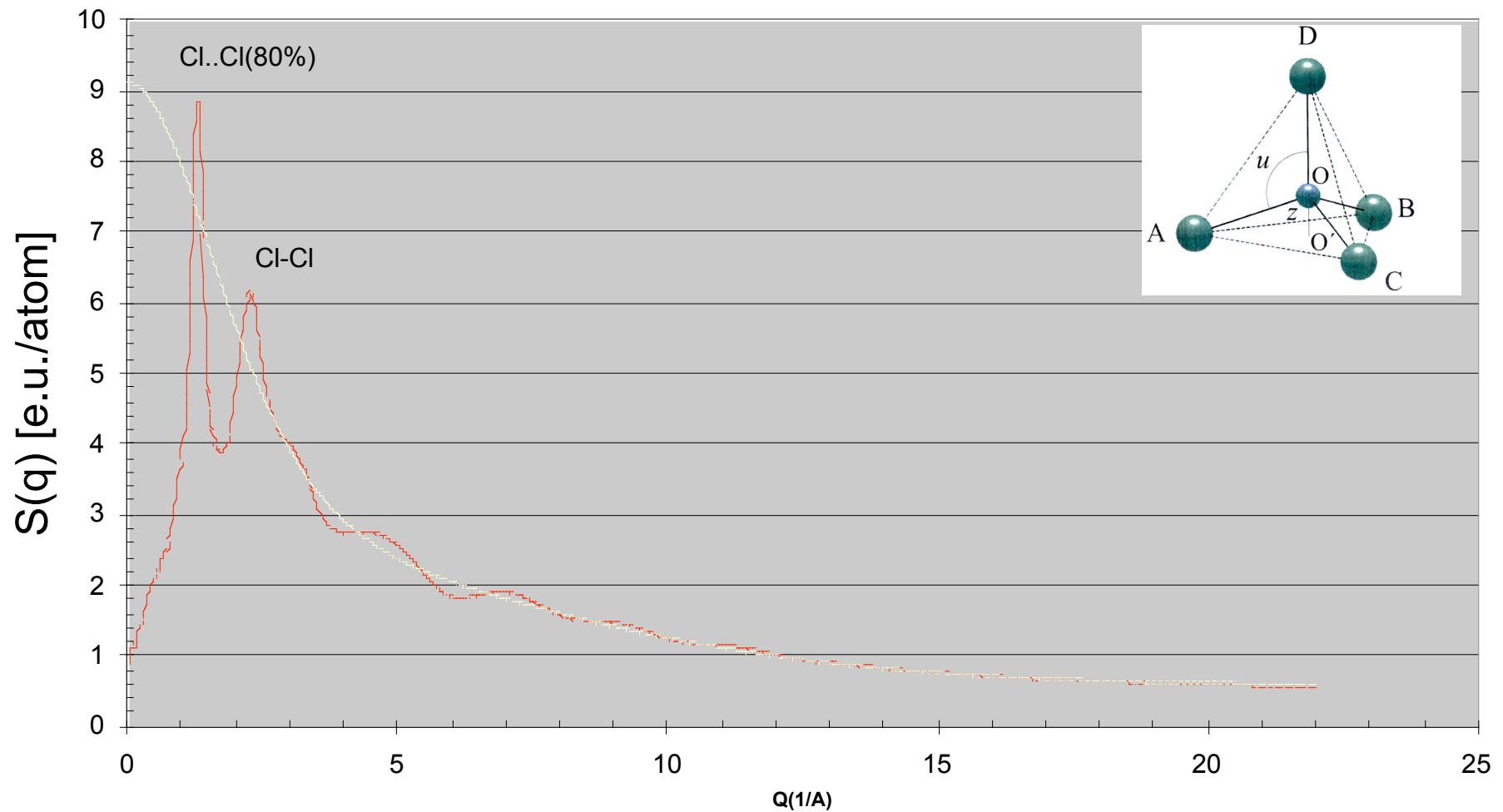
$\Delta S[r, t]$ is a formfactor biased measure of the change in the electron density seen from the center of an average excited atom.

The $g(r)$ function in a liquid

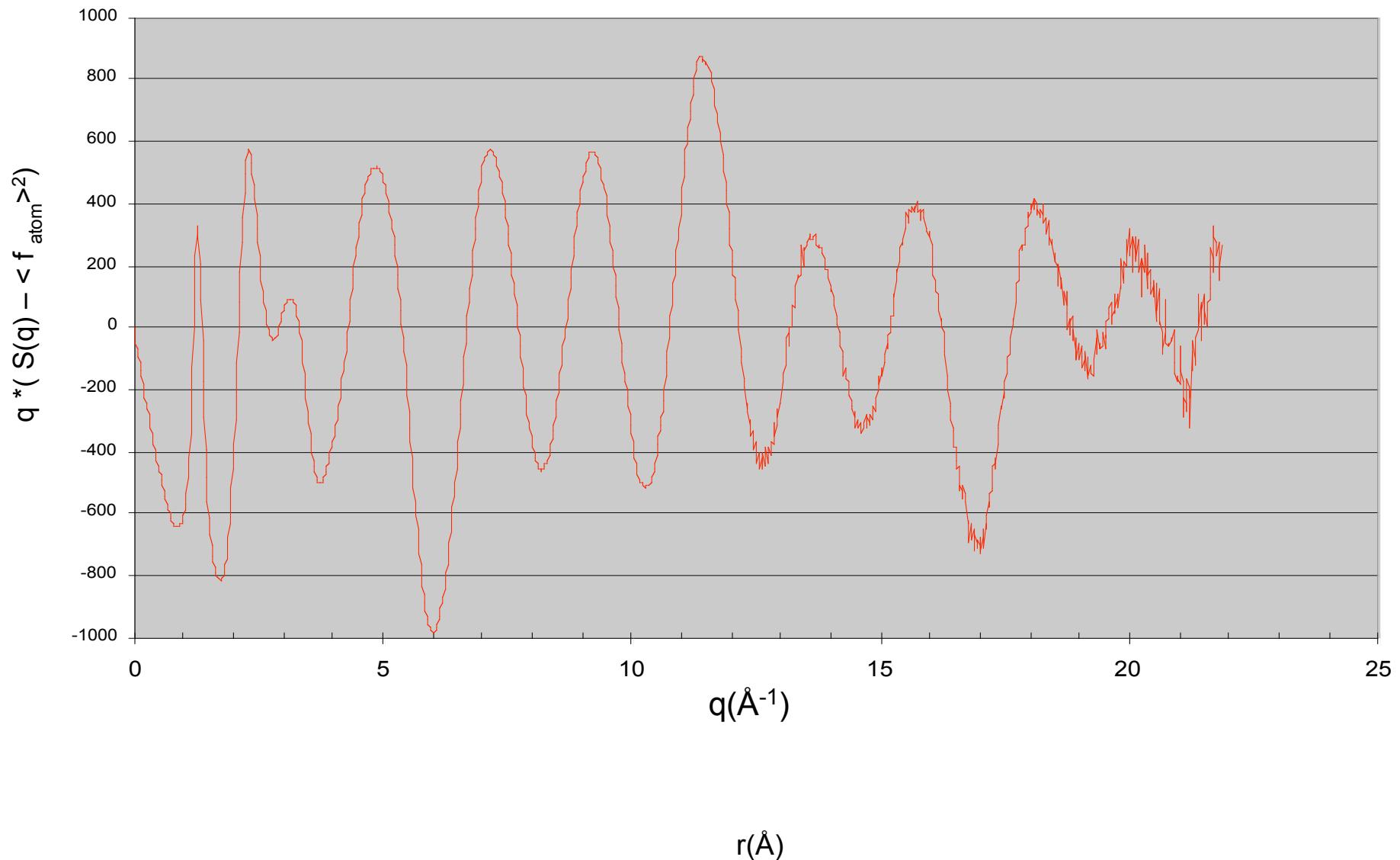


The Radial Diffraction Pattern from CCl₄

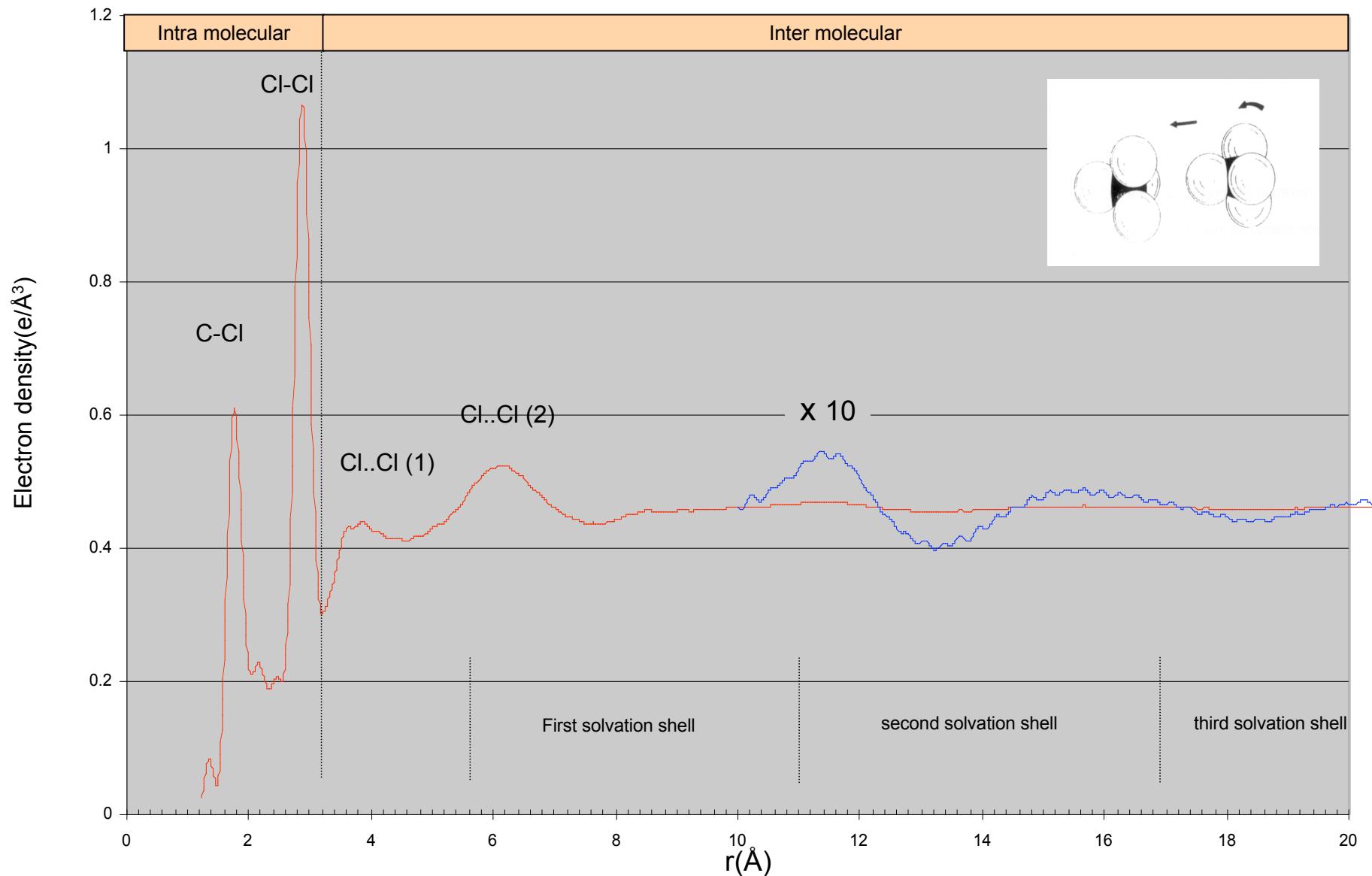
E= 88.0 keV, $\lambda= 0.141 \text{ \AA}$, exposure time 12 s on ID15(ESRF)



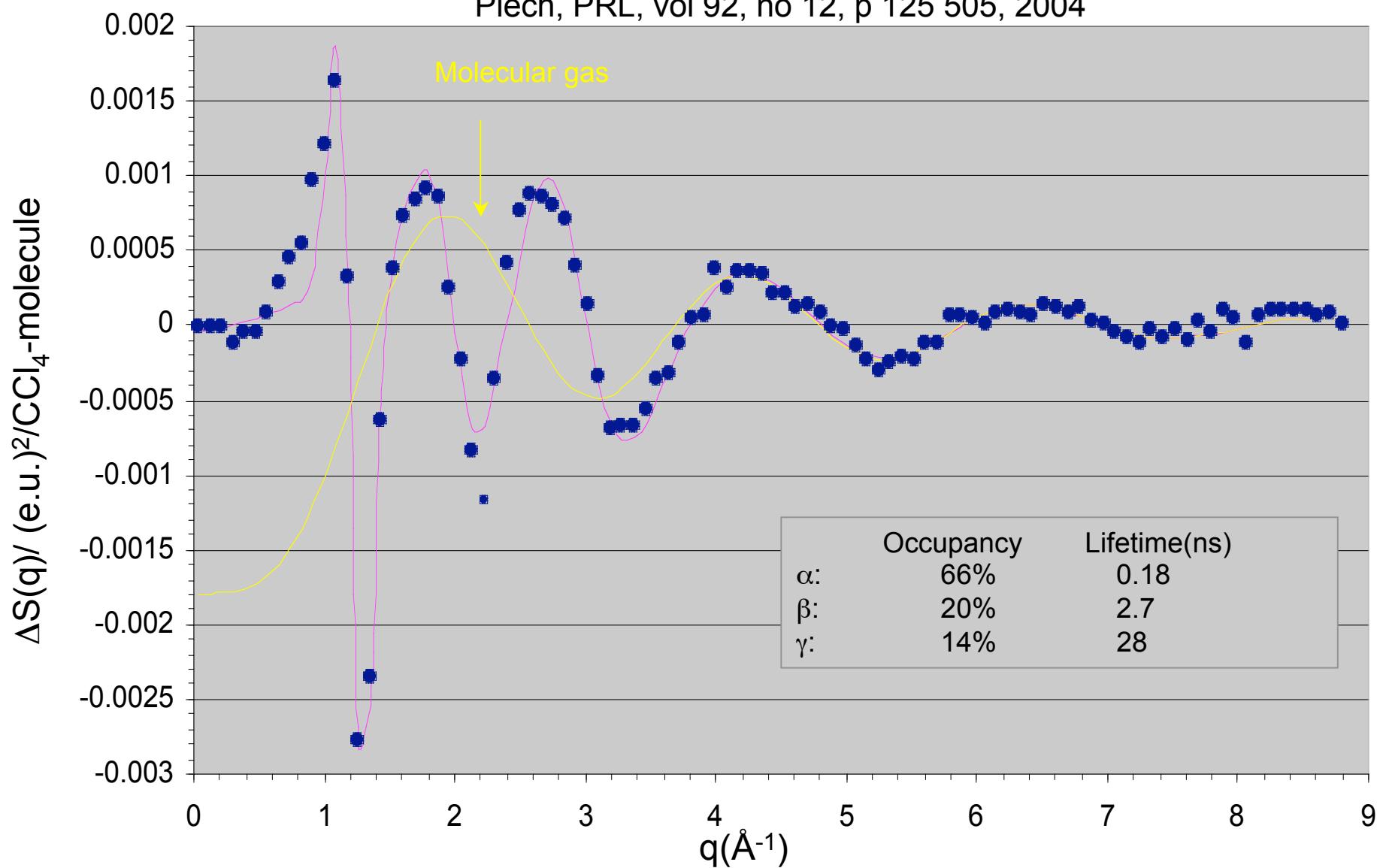
The interference from local order in CCl_4
static measurement from ID15, ESRF.



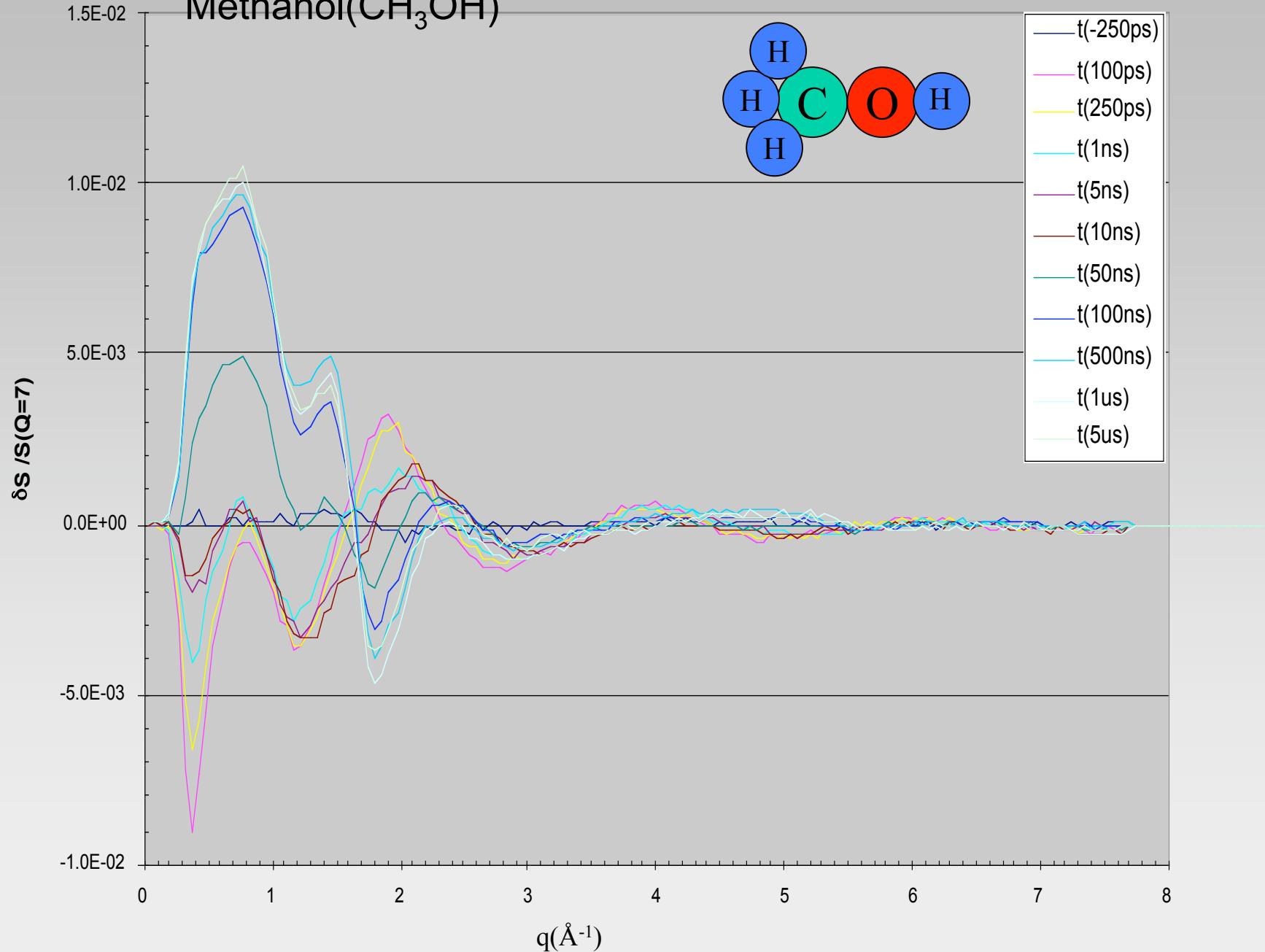
The measured structure of liquid CCl_4



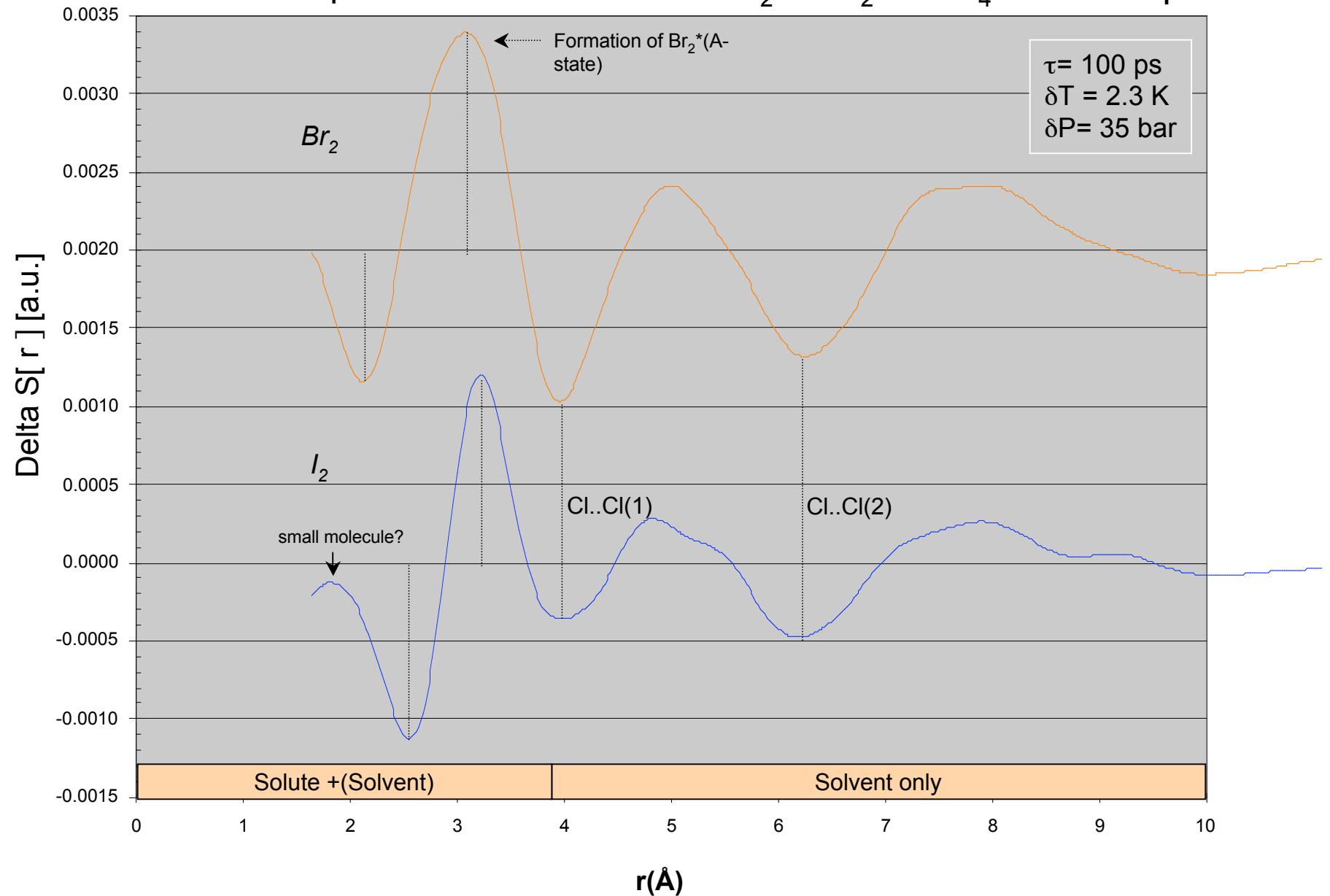
The difference oscillations from $I_2^* + CCl_4^*$ 100 ps after excitation.
Plech, PRL, vol 92, no 12, p 125 505, 2004



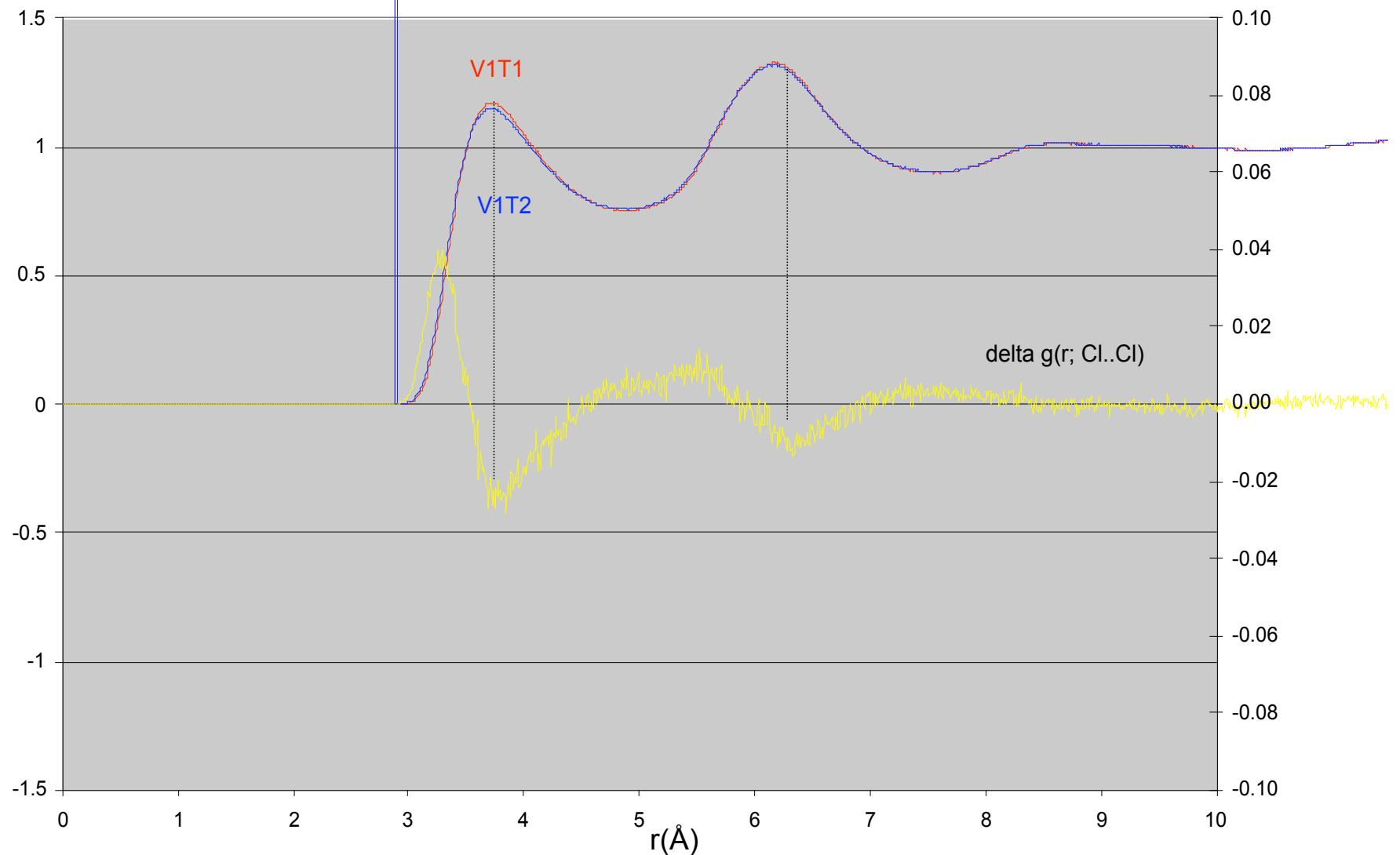
Difference oscillations from I_2^* in Methanol(CH_3OH)



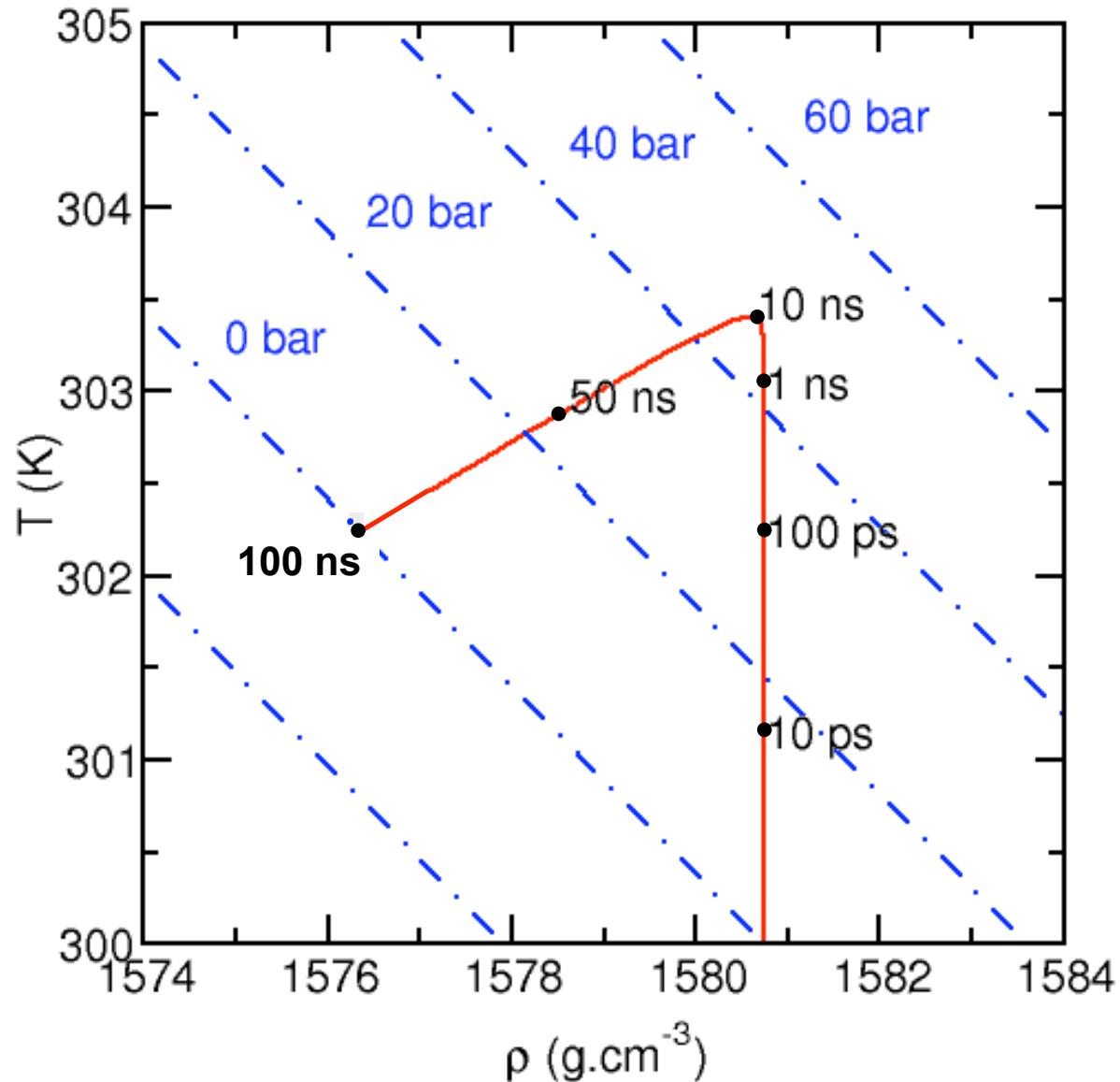
Radial map of the recombination of Br_2 and I_2 in CCl_4 after 100 ps



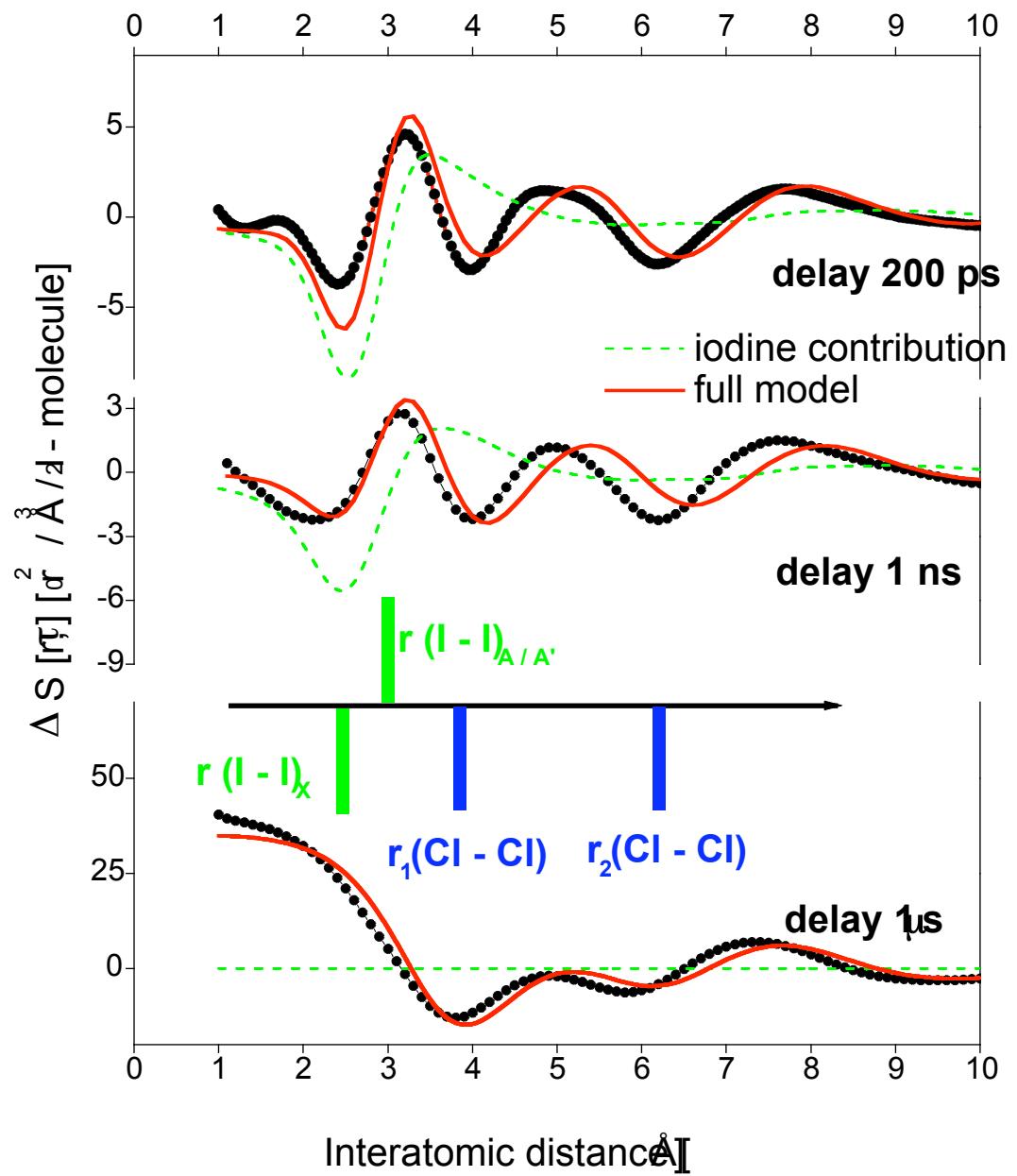
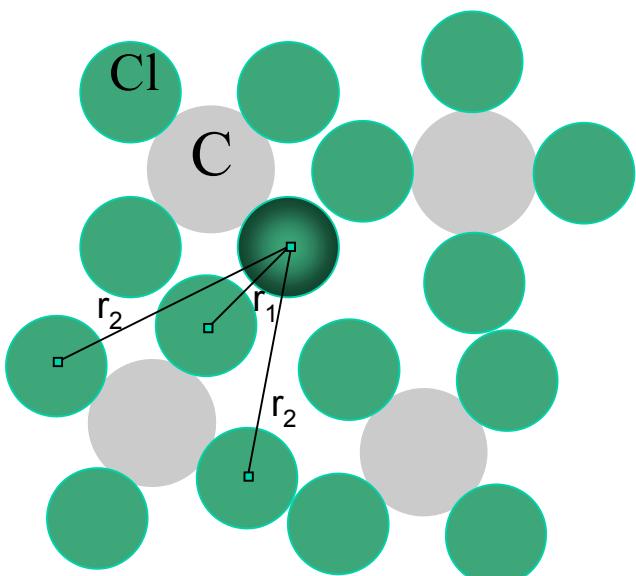
The effect of an ultrafast temperature jump on $g_{\text{ClCl}}(r)$.
MD simulation.



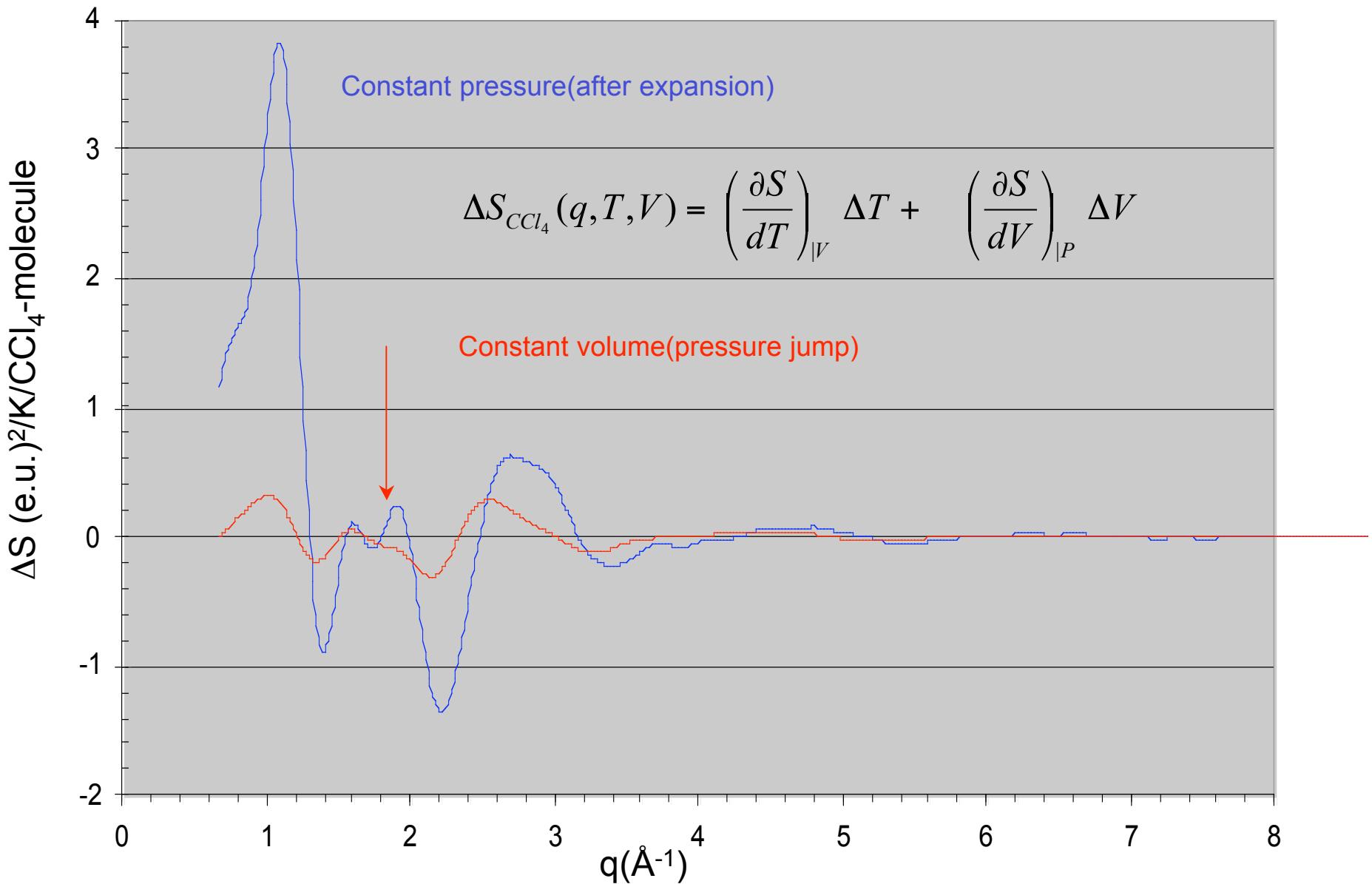
The energy transfer $I_2^* \rightarrow CCl_4$ causes a change in the thermodynamic state of bulk CCl_4

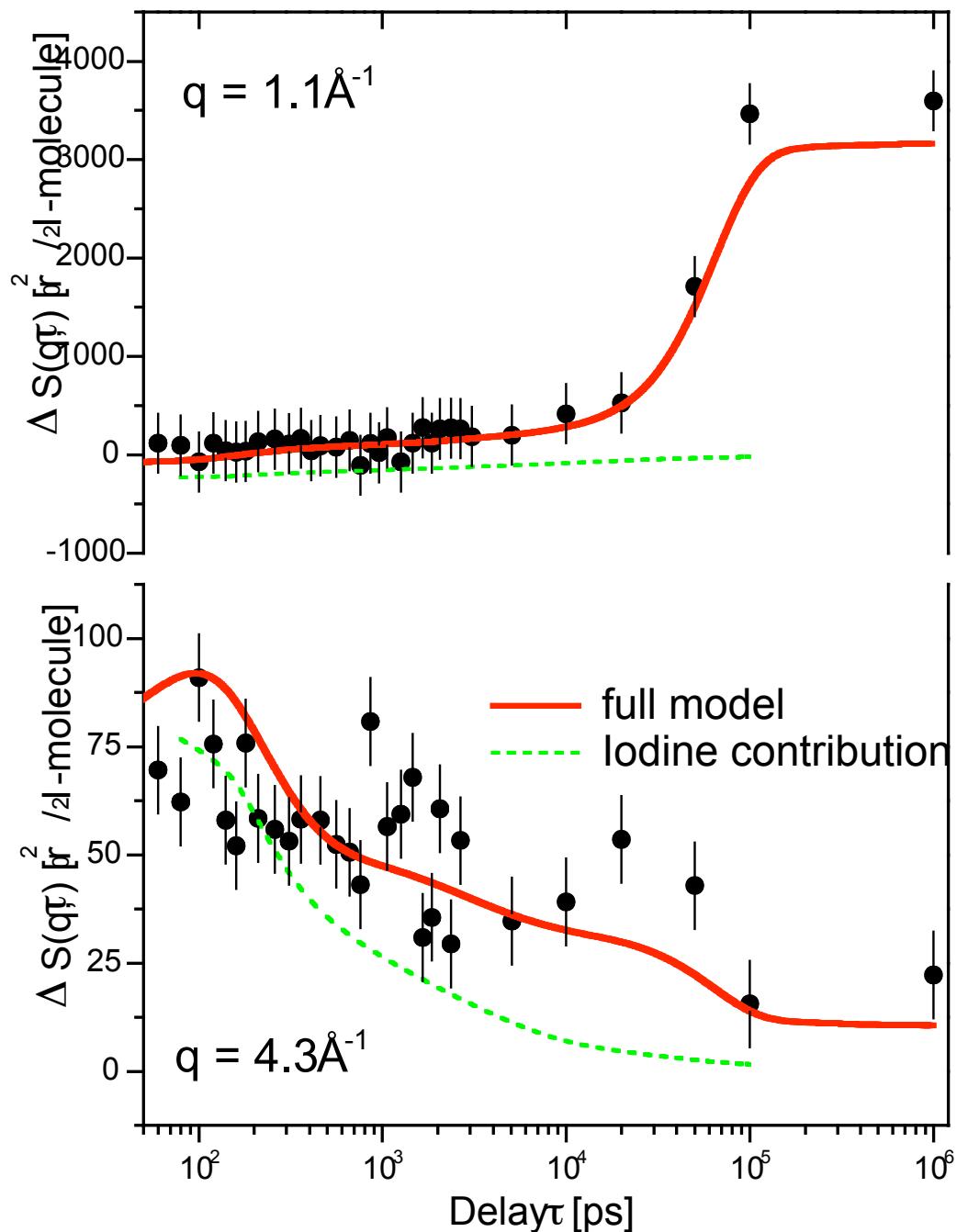


Dynamics in real-space

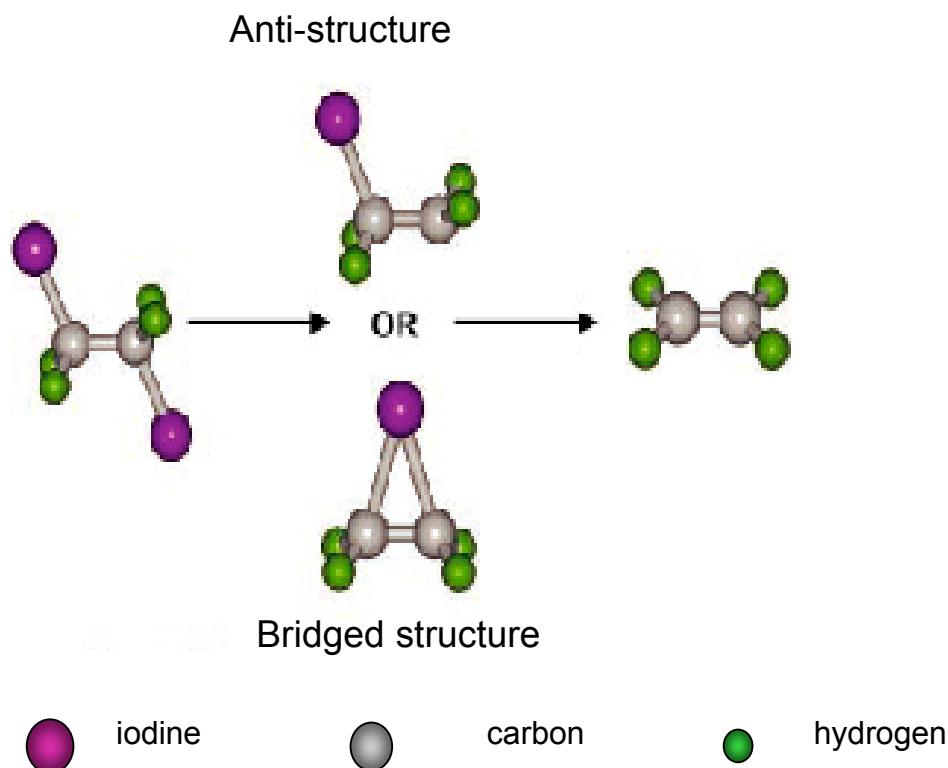
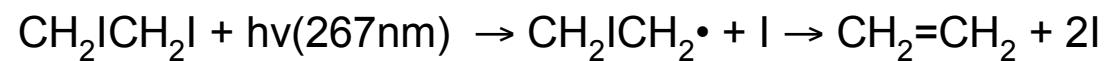


The signals from the induced change in structure of CCl_4

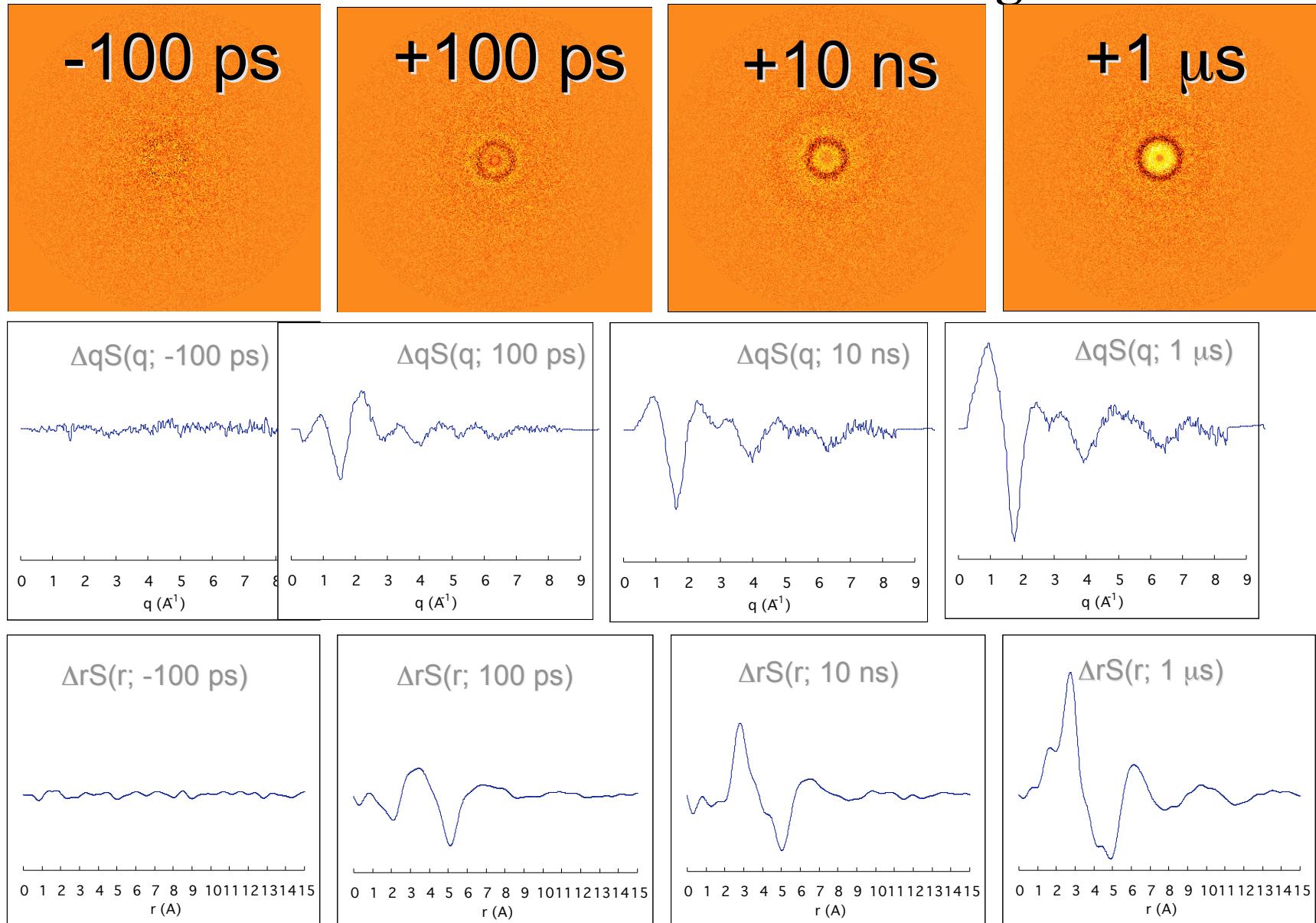




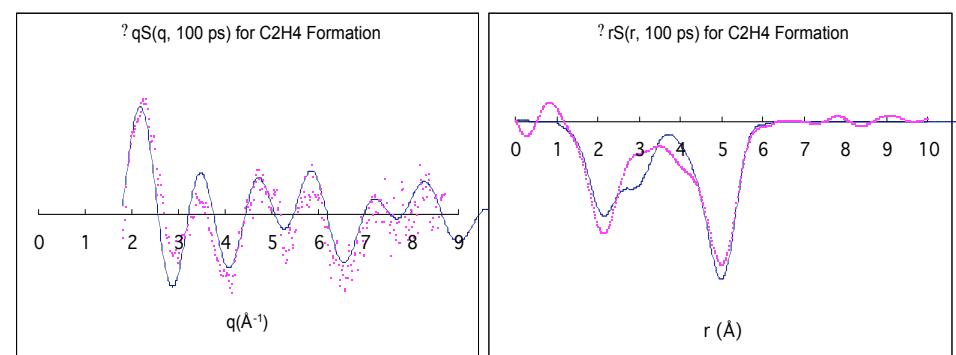
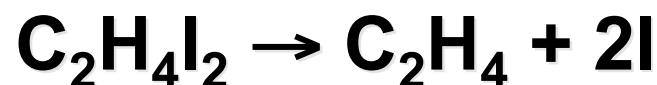
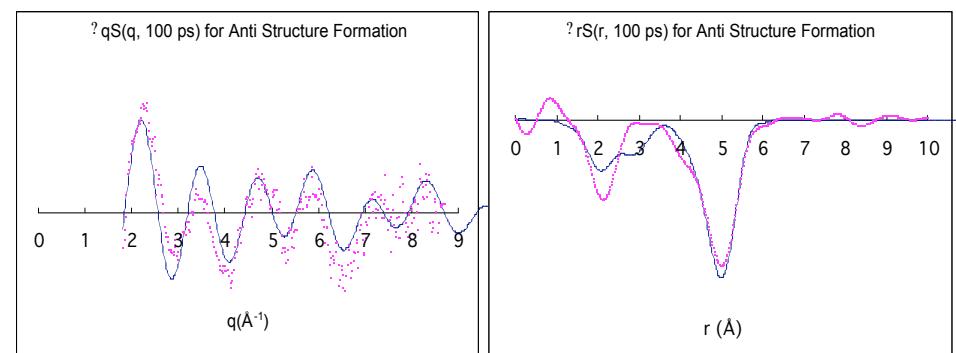
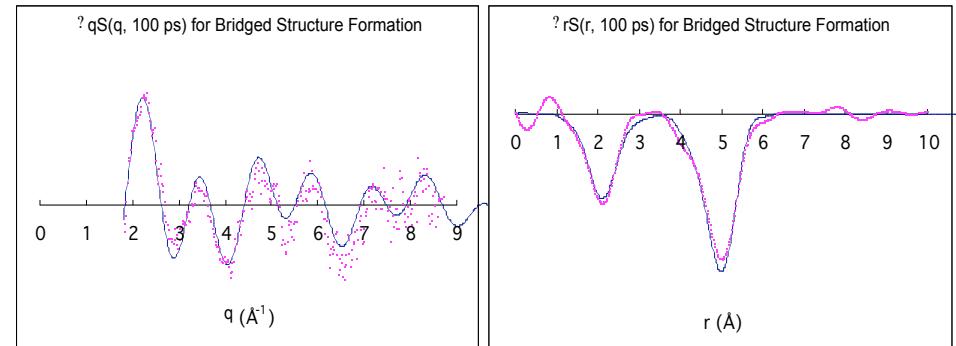
The Diodoethane project(Hyotcherl Ihee et al.)



Time-Resolved Difference Signal



Molecular Structure of $\text{C}_2\text{H}_4\text{I}_2$ intermediate at 100 ps



The bridged structure is formed!

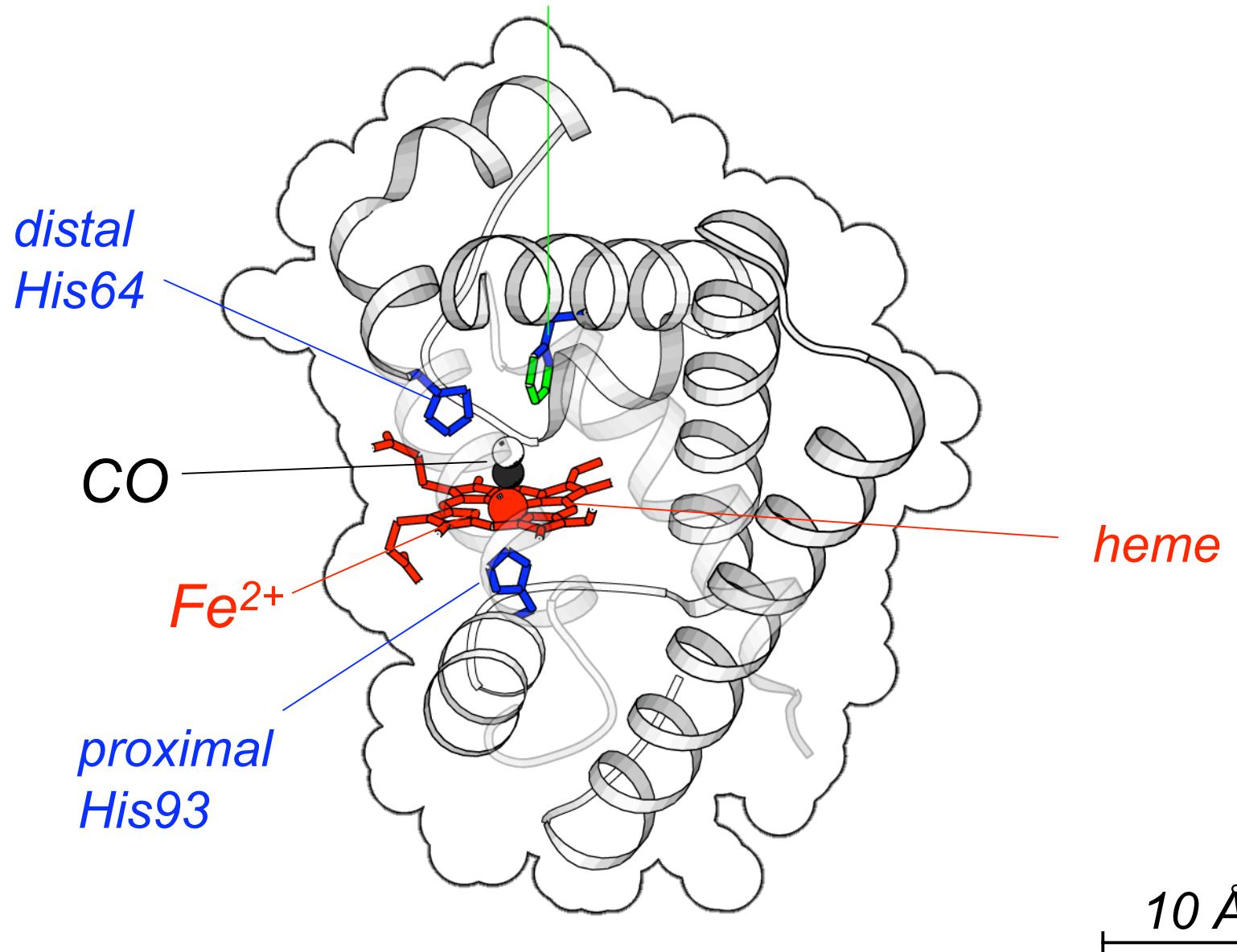
The time-resolved structure of the myoglobin complex MbCO

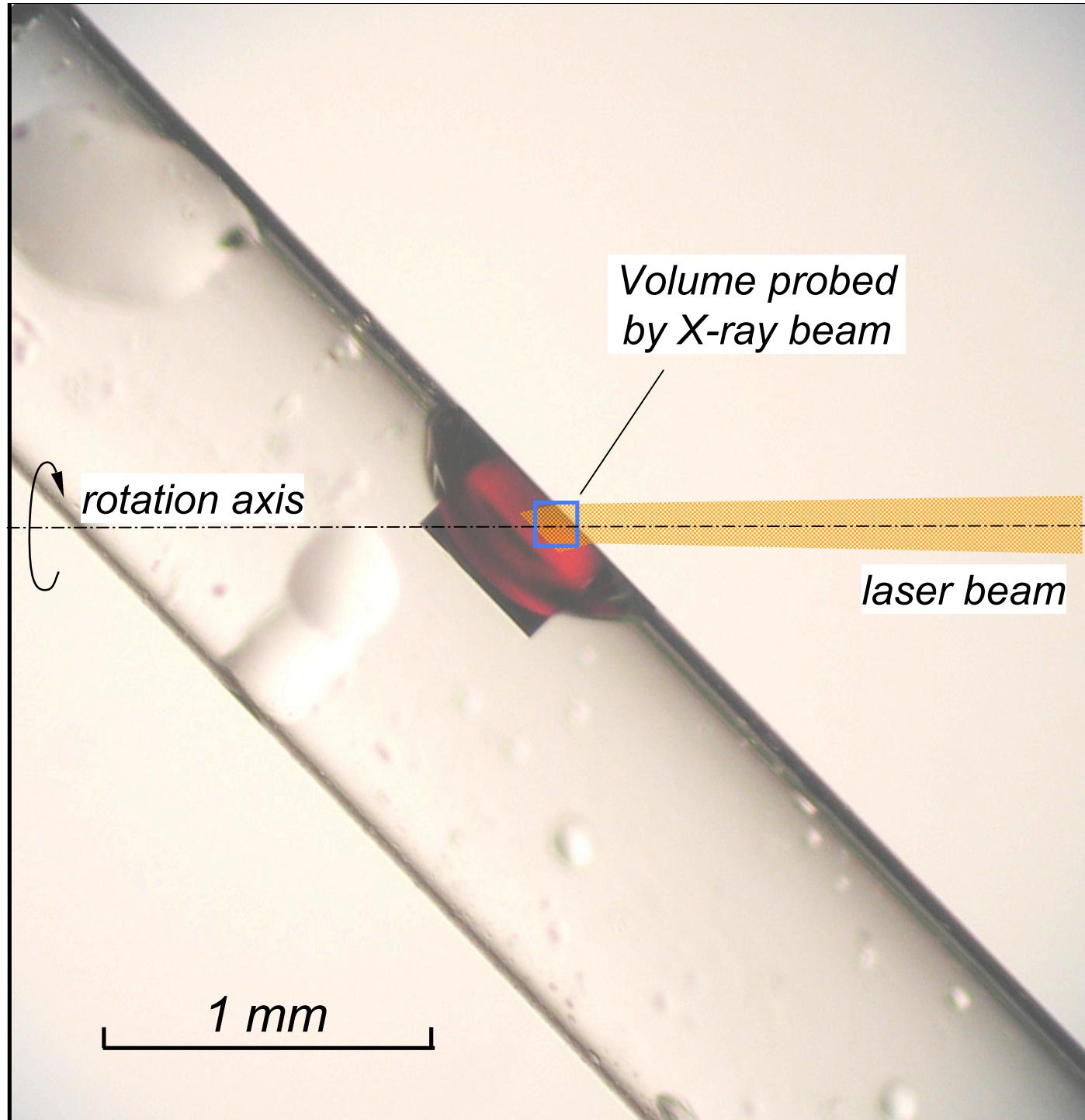
Team:

Friedrich Schotte Philip Anfinrud, Michael Wulff, Dominique Bourgeois,

Myoglobin-CO L29F

Phe29

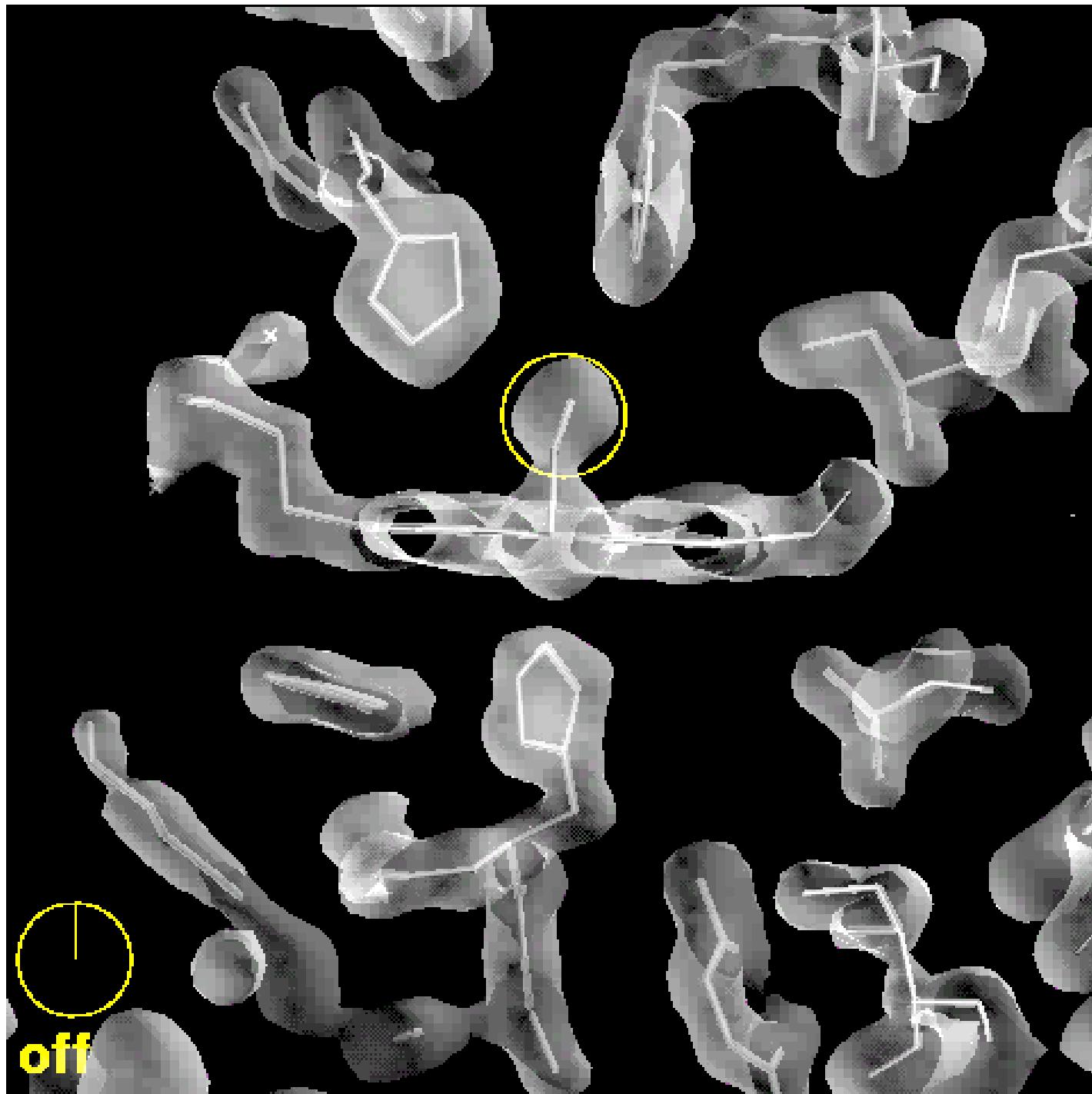




Laue diffraction image

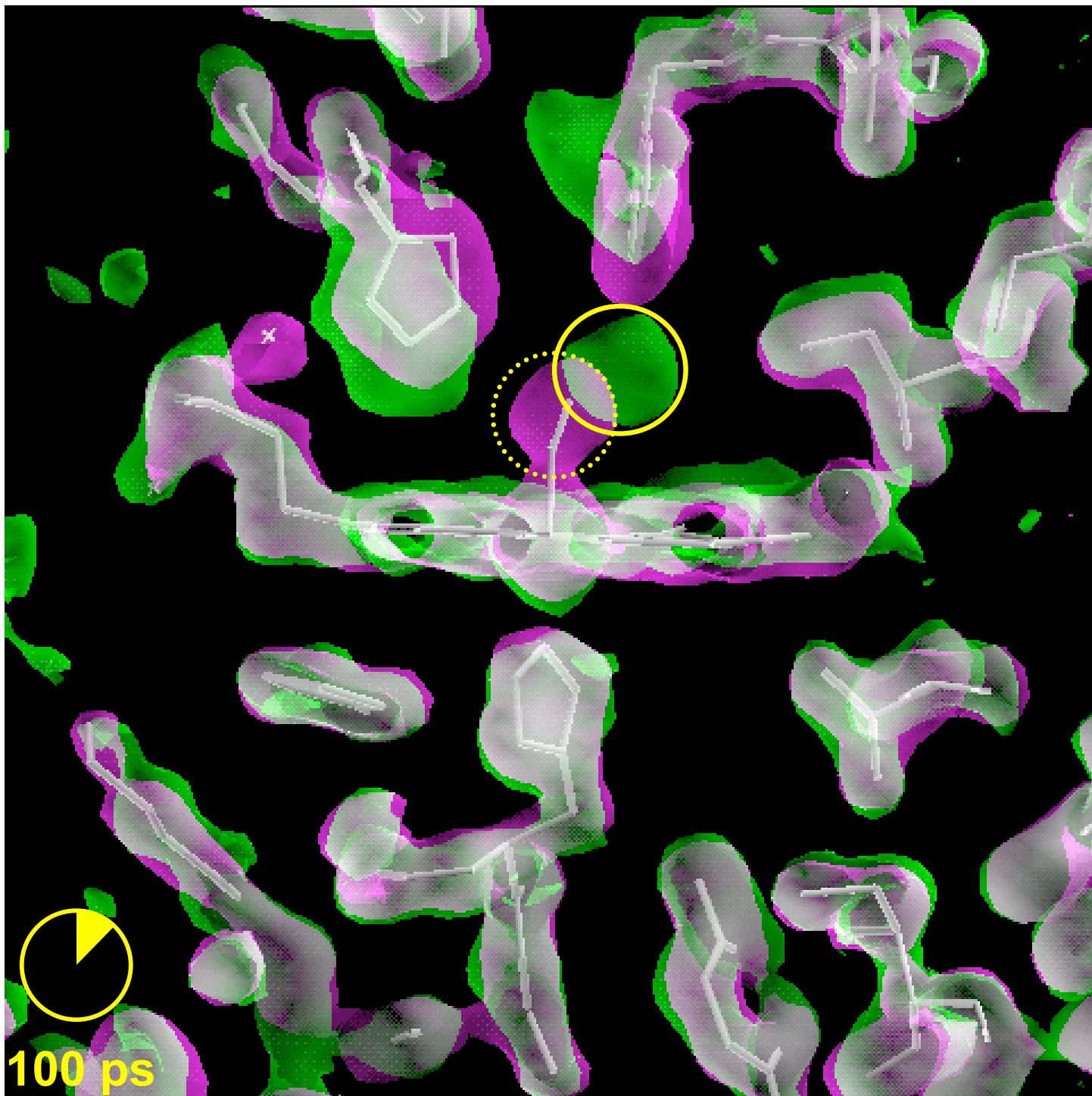
of 150 ps
photolysis

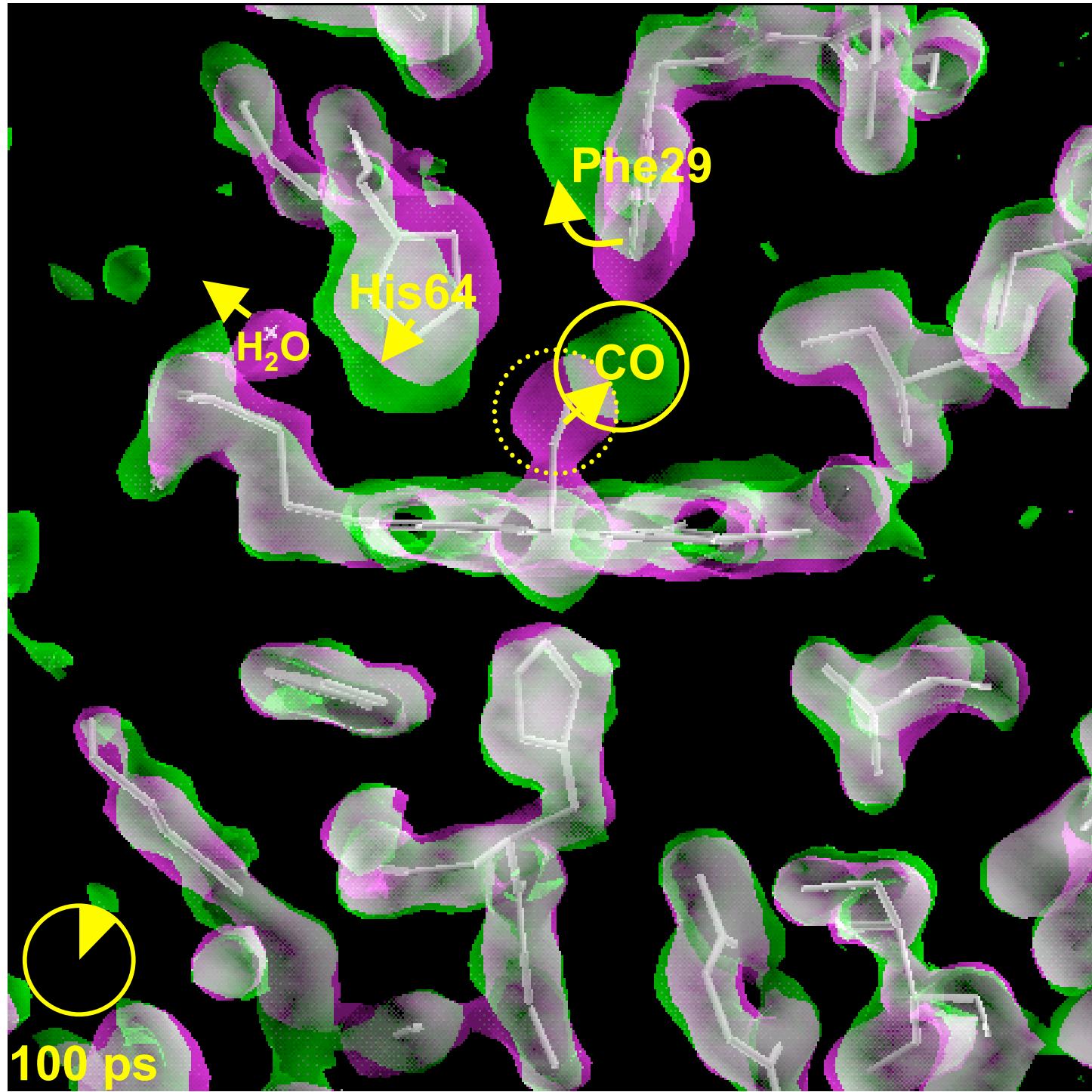
L29F12_1

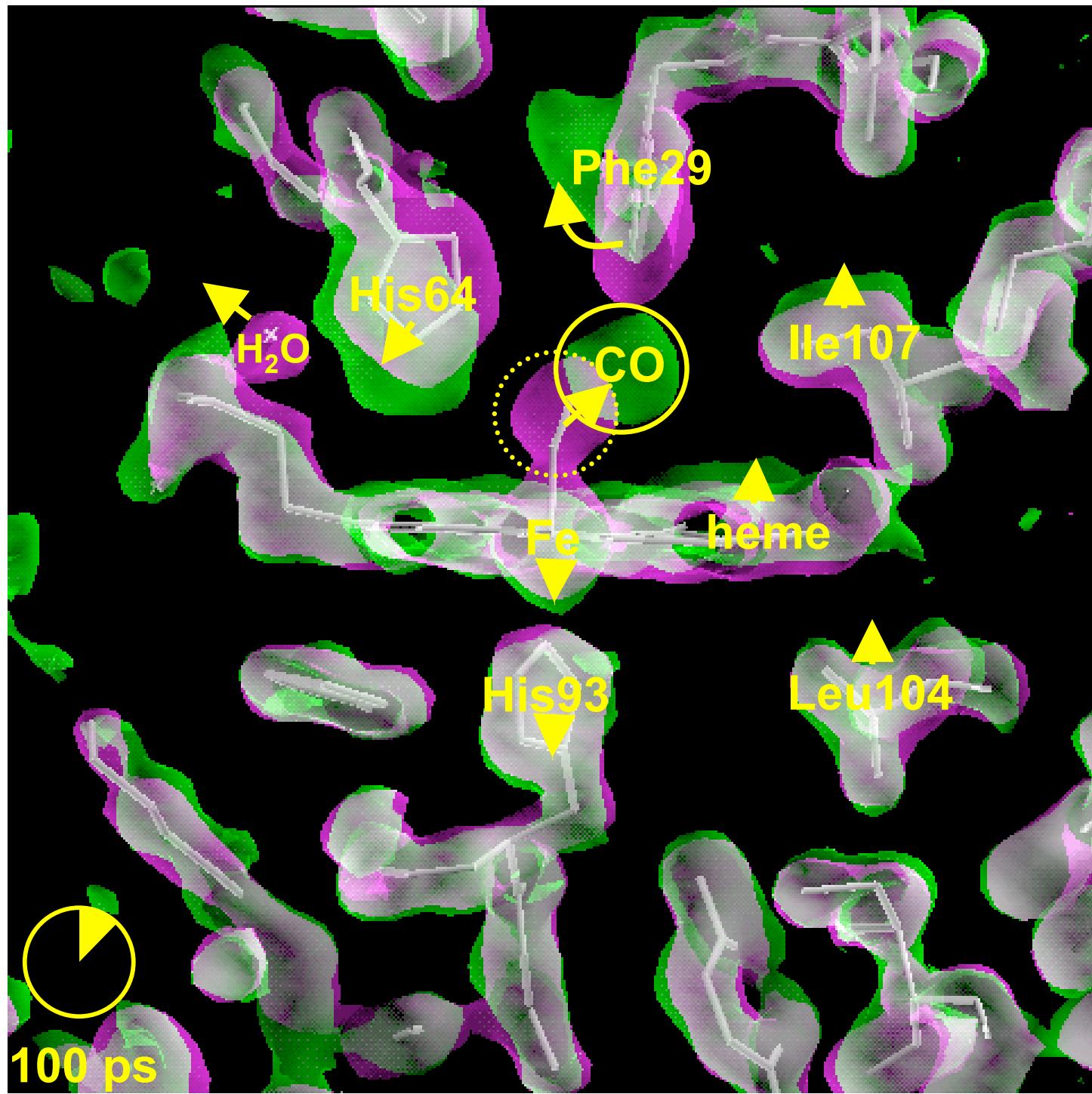


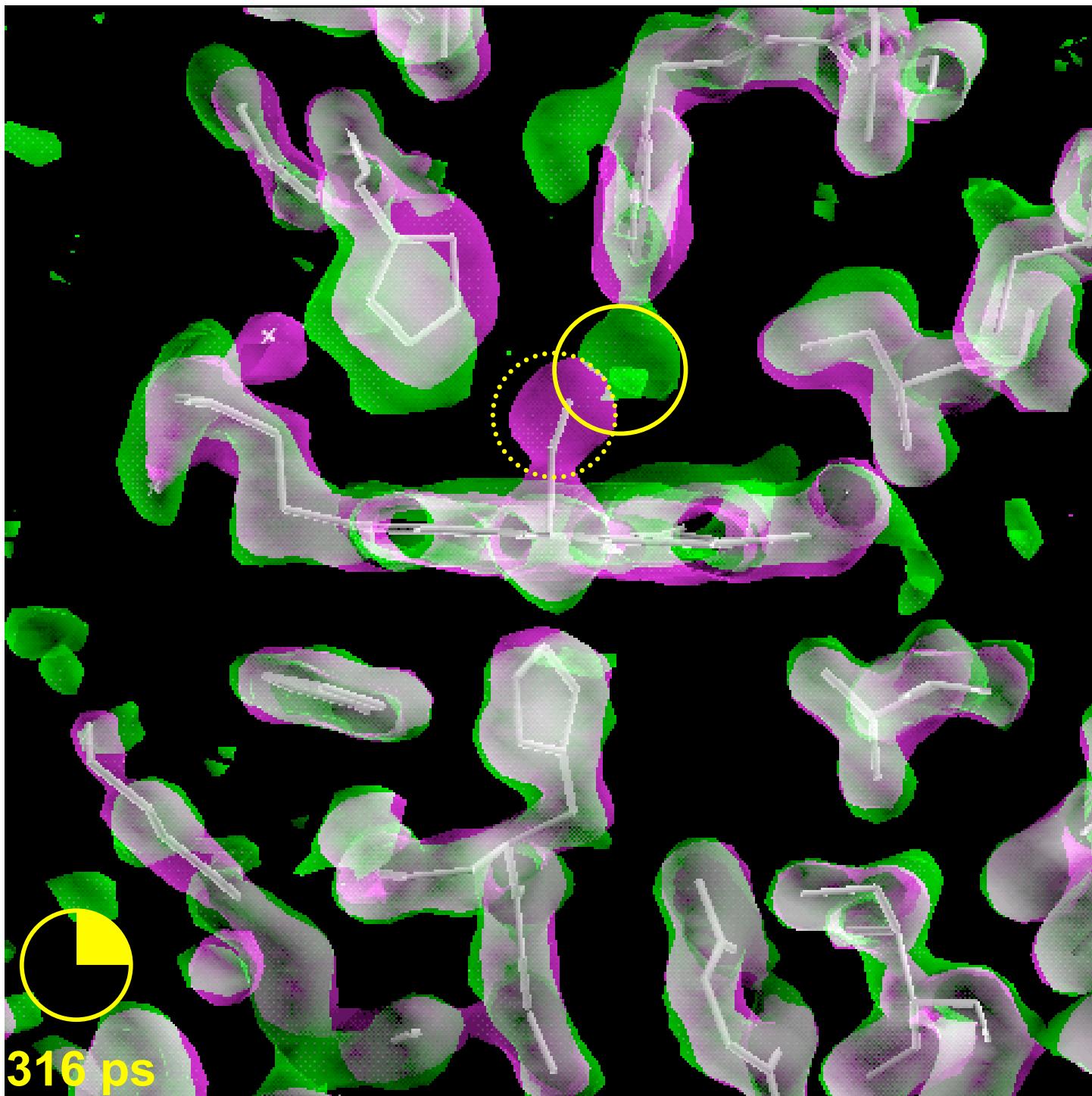
off

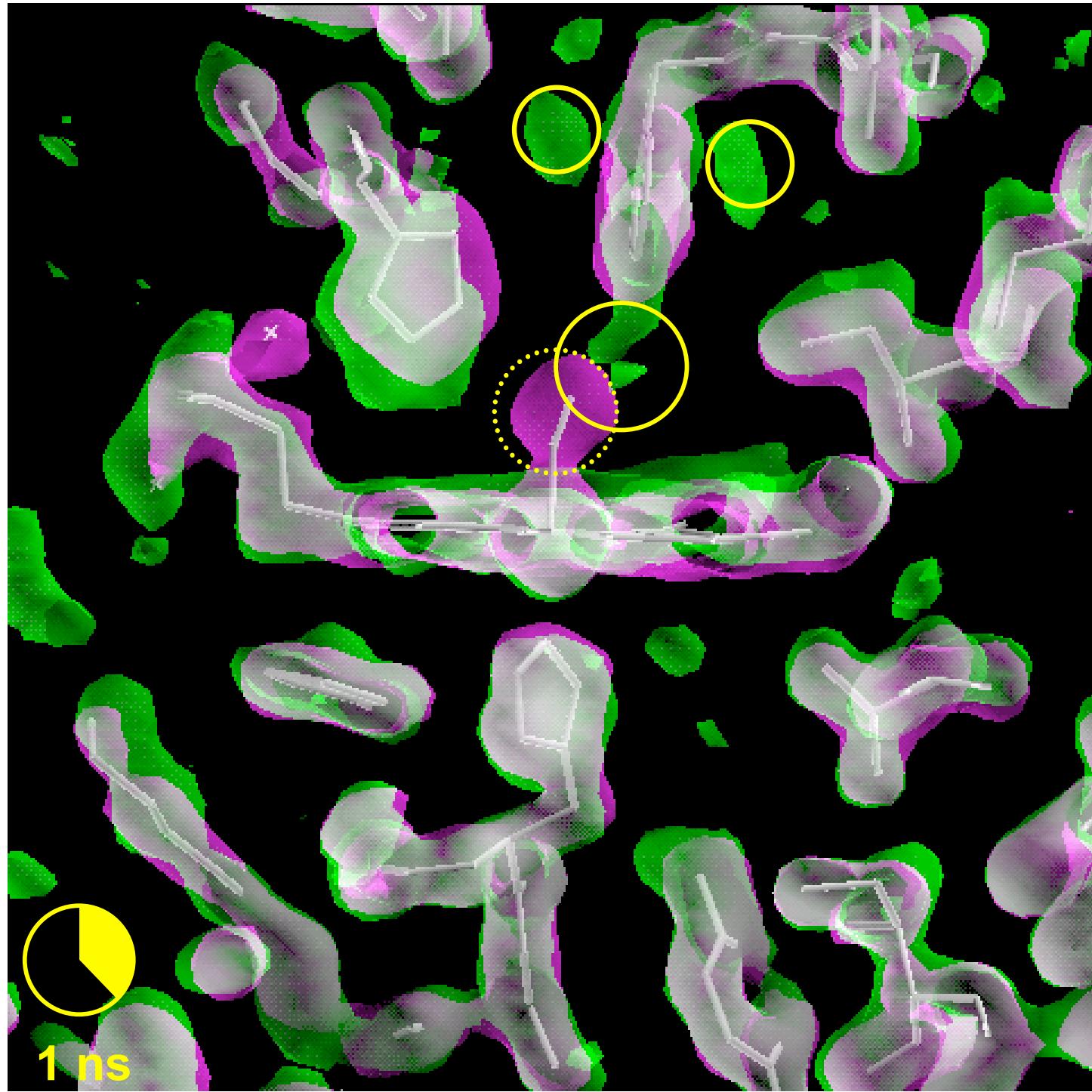
Science, 300, 194-
(20 Jun 2003)

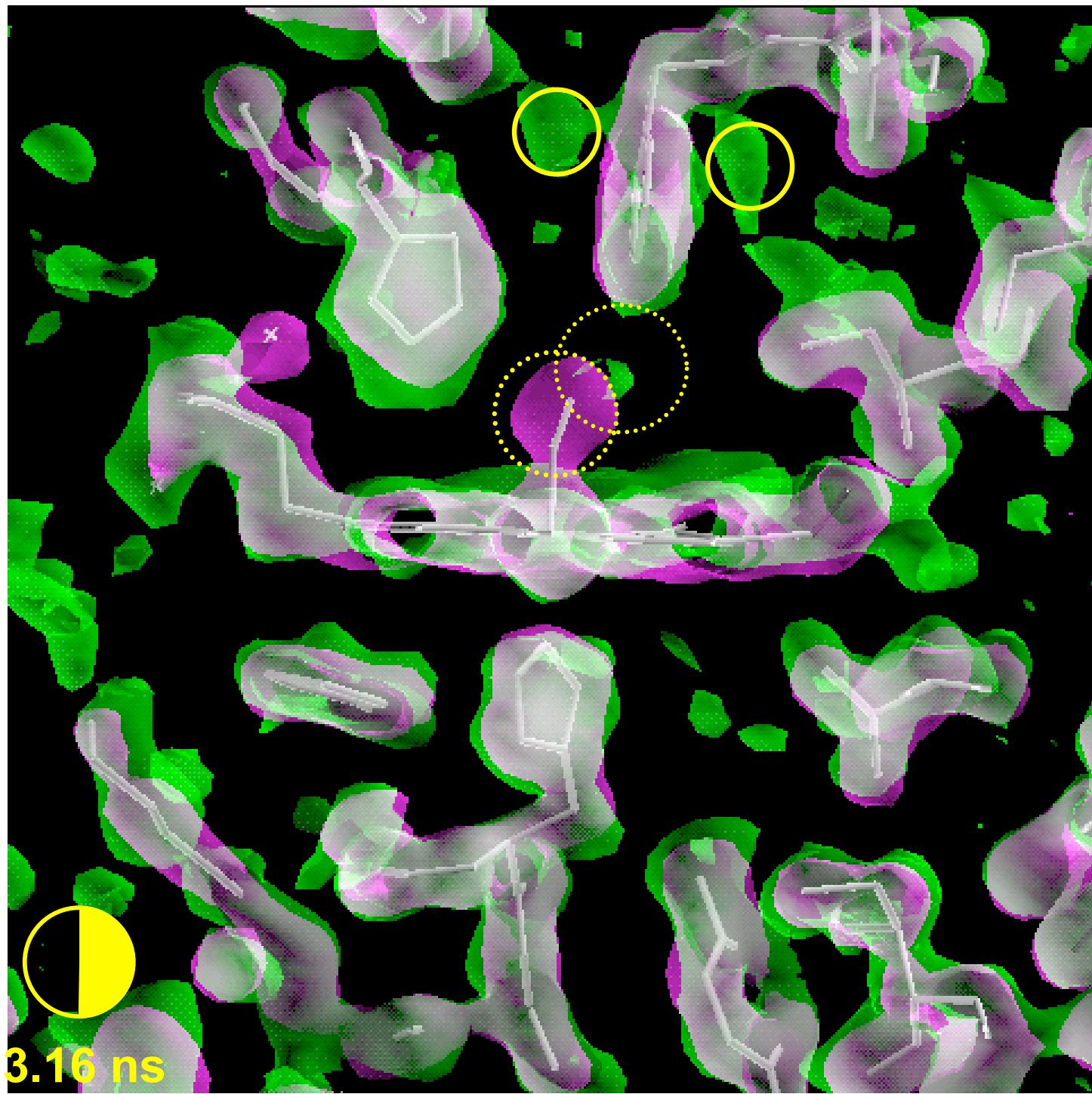


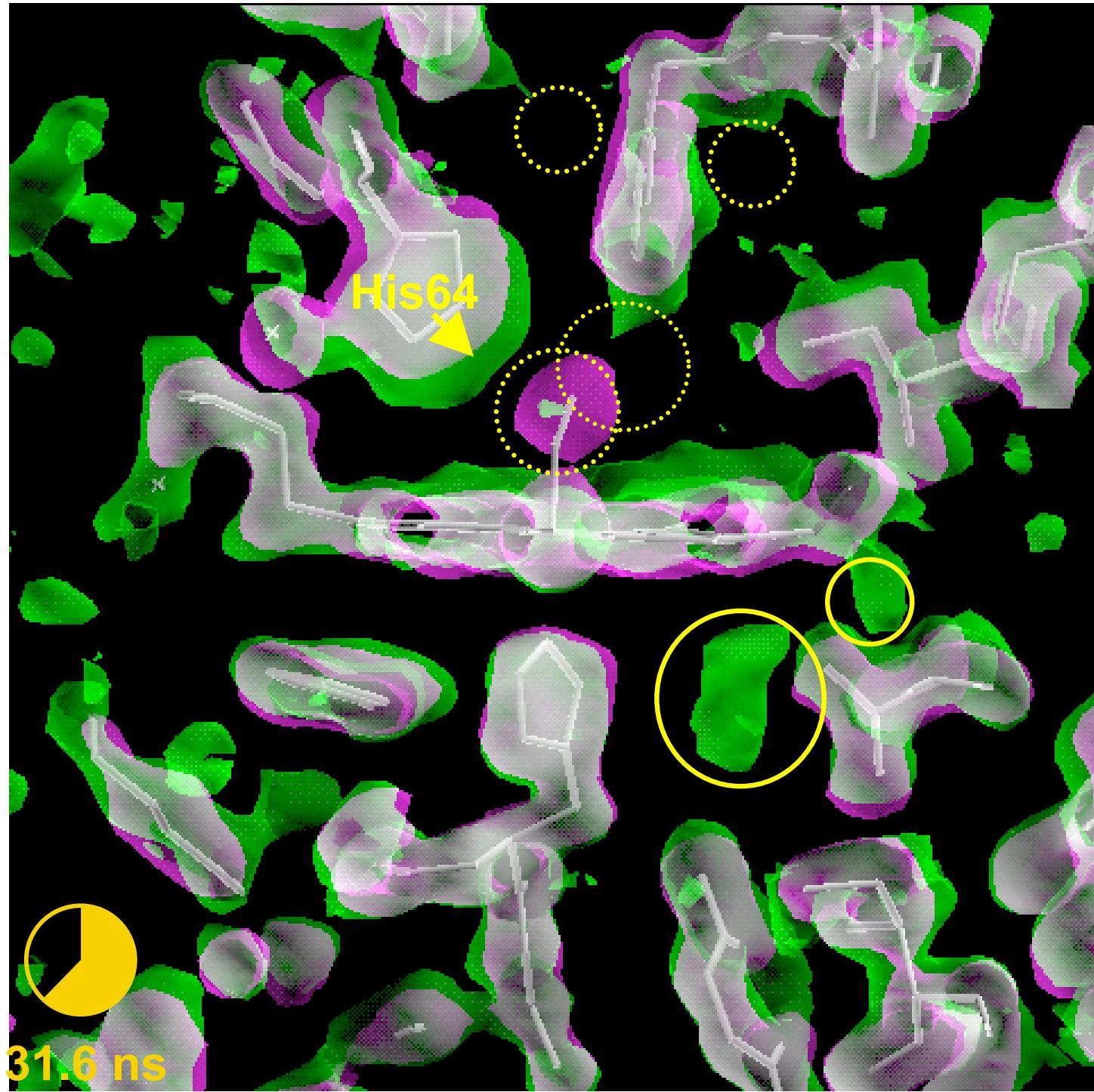


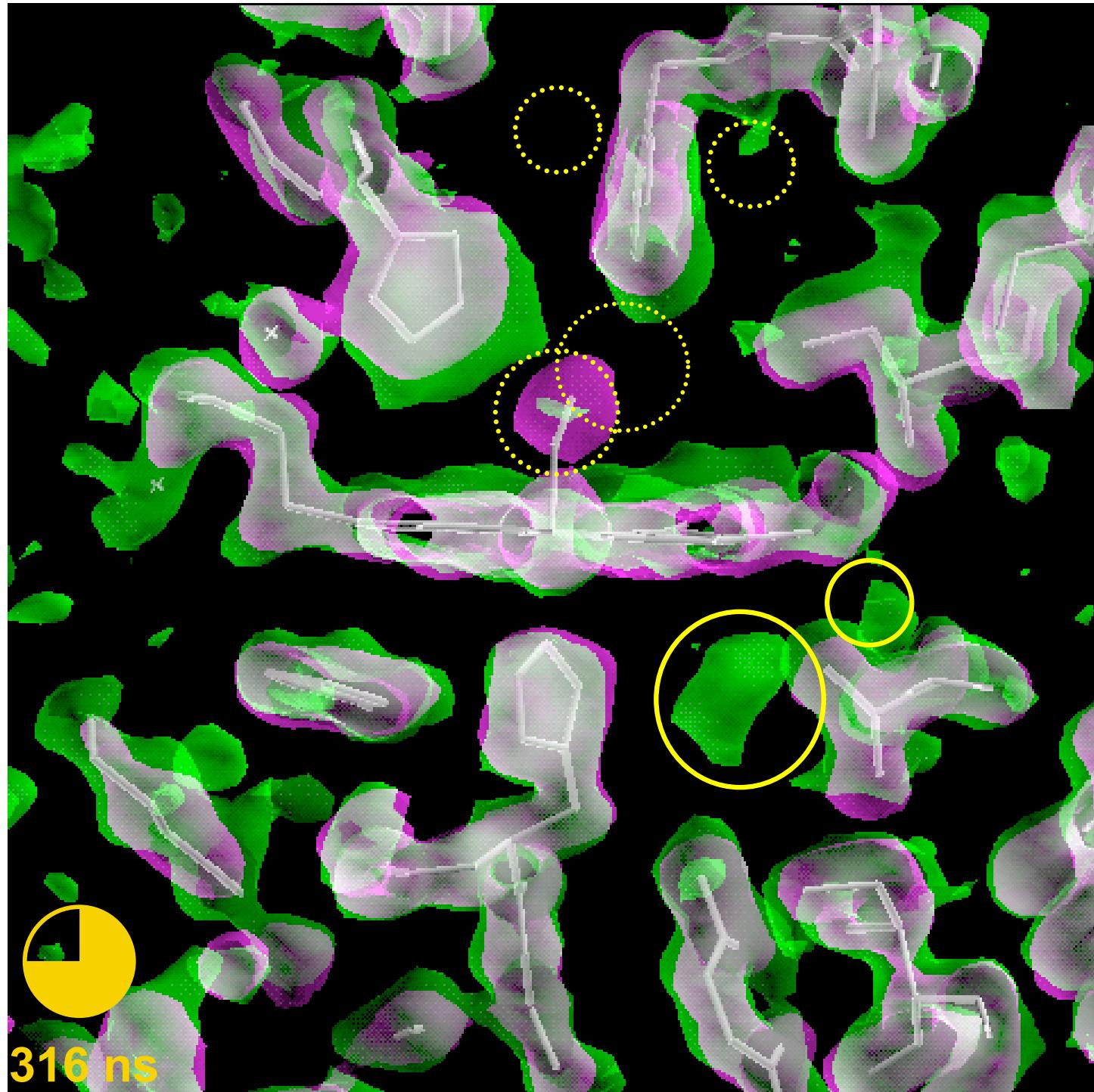


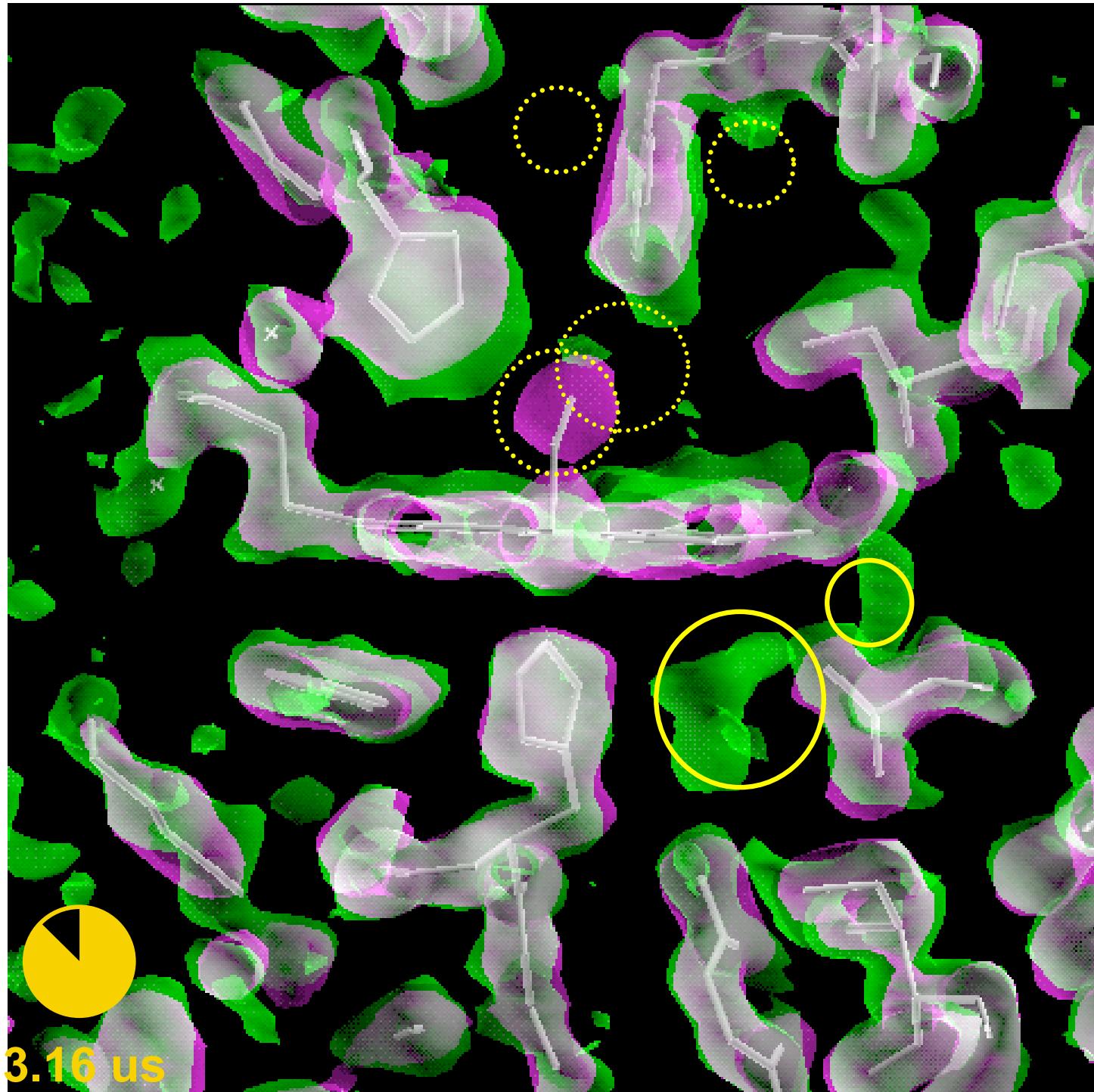


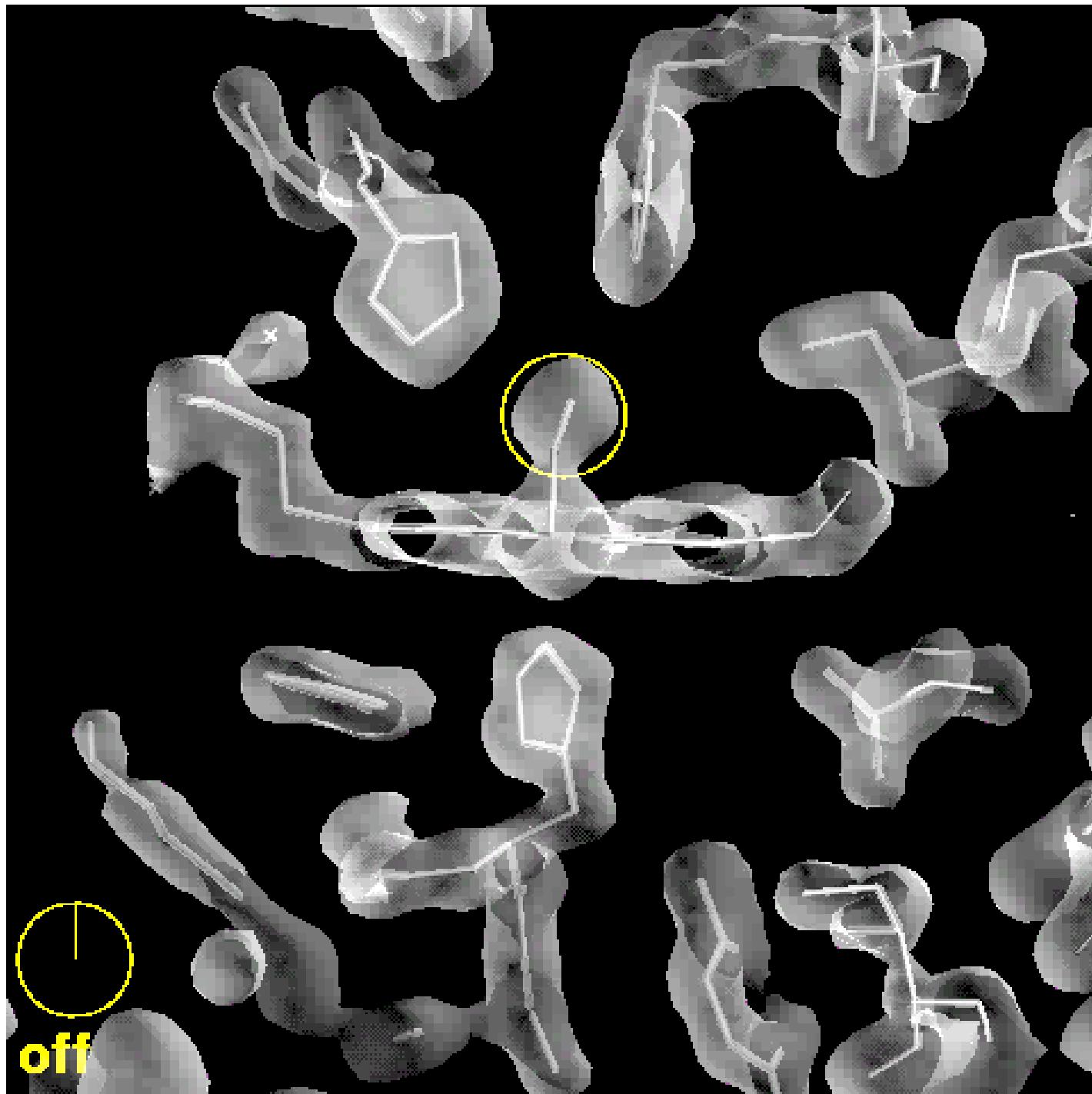












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(20 Jun 2003)

Outlook

1. Increase the q-range from 0.5-9.0 to 0.05-13.0(ML optics and new U20 undulator)
2. Use multilayer optics for better spectral control(and lower heatload)
3. Use hybrid mode to reach 50 ps resolution
4. Use picosecond laser pulses to excite proteins
5. Have a faster CCD camera to increase the x-ray duty cycle from 50% tp 90%

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